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A STUDY TO EVALUATE TECHNIQUES OF ASSESSING AIR POLLUTION DAMAGE TO PAINTS

G. G. Campbell, et al

Sherwin-Williams Company

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FINAL REPORT

A STUDY to EVALUATE TECHNIQUES of ASSESSING AIR POLLUTION DAMAGE to PAINTS

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FINAL REPORT

on

A Study to Evaluate Techniques of Assessing Air Pollution Damage to Paints

to

Division of Ecological Research
Materials Branch
Environmental Protection Agency

Contract No. 68-02-0030

May, 1977

by

G. G. Campbell, G. G. Schurr and
D. E. Slawikowski

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FOREWORD

This report was prepared by The Sherwin-Williams Company under Contract No. 68-02-0040 "A Study to Evaluate Techniques of Assessing Air Pollution Damage to Paints" for the Environmental Protection Agency, Division of Ecological Research, Materials Branch, Research Triangle Park, North Carolina with Mr. J. W. Spence as Project Officer.

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II

ABSTRACT

Erosion rate studies supported by ATR measurements and SEM analyses provide a definitive technique for determining the effect of atmospheric pollutants on the performance of exterior coatings. This technique is based on the use of a gas controlled, Xenon Arc type Weather-Ometer. The other methods of testing that were investigated - tensile strength, gloss or sheen and surface roughness - provided considerably less consistent trends in data for either the "short term" exterior exposure or the Weather-Ometer studies.

Atmospheric pollutants at levels representative of a highly polluted industrial site (1.0 ppm SO_2 or O_3) were shown in Weather-Ometer studies to exert a significant adverse effect on the performance of specific coatings compared to the zero pollutant condition. In general, 1.0 ppm SO_2 as compared to the zero pollutant level caused a considerable effect on the oil house paint, a moderate effect on the latex and coil coatings, but no effect on the alkyd industrial maintenance paint and the automotive refinish. Based on the comparison of erosion rates, SO_2 at the 1 ppm level affected these coatings to a greater extent in either the shaded or unshaded condition than exposure to 1 ppm O_3. Graphs of the erosion rates with accompaning 95 percent confidence limits versus pollutant level (zero, 0.1 and 1 ppm SO_2 or O_3) supports the a priori hypothesis that erosion rates are linearly related to pollutant concentration. In addition, the ranking of a coating in terms of erosion rates was also shown to be virtually independent of exposure to a pollutant type or level.

The erosion rates for coatings exposed at the Chicago and Valparaiso exterior testing sites were higher than at the Los Angeles and North Dakota locations. This trend in data is explained by the higher SO_2 level at the two former exposure sites. ATR measurements and SEM photomicrographs also support this contention. In addition, the ranking in terms of mils loss was virtually the same as the erosion rates generated in the Weather-Ometer studies regardless of exposure location (both north and south).

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I Introduction

Only during recent years has there been any serious attention focused on the potential adverse effects that various air pollutants (particularly ozone and oxides of sulfur) may have on polymeric materials.[1] Most published studies of a fundamental nature have been concentrated in the areas of rubber,[2] textiles,[3] and plastics.[2,4] Very few of these investigations have concerned the influence of air pollutants on the performance of exterior coatings,[5,6] and these were conducted at excessively high pollutant levels compared to normal exterior conditions. They showed that pollutants, specifically SO_2, exert a detrimental effect on coating properties and components.

Evaluation programs of coating products designated for exterior use typically involve environmental exposure at a number of locations. Attempts to pinpoint the reason(s) behind unusual performance behavior and particularly, ascribing these effects to atmospheric pollutants are obviously difficult because so many factors are involved simultaneously during exterior weathering. However, there have been several instances where air pollutants were suspected of promoting premature film failure, notably "hazing" or "frosting" and intercoat peeling. The former problem has been observed on the surface of specific coatings (readily detectable with colored systems) exposed in polluted environments particularly containing moderately high levels of SO_2. The objectionable surface deposits noted especially under eaves where moisture condensation is apt to occur would normally require repainting by the customer. If the repaint surface is not cleaned properly, intercoat peeling could result.

A severe problem of intercoat peeling in protected areas under the eaves also has been related to the formation of water sensitive materials from zinc oxide containing repaint surfaces.[7] It is interesting to note that a 1960 survey conducted in approximately 100 areas in the United States showed that the most consistent reports of severe intercoat peeling came from the Great Lakes region. The generally recognized high SO_2 level associated with this industrially oriented area could cause formation of the water soluble zinc sulphate and subsequent adhesion problems.

A study concerning the corrosion of steel provided relatively conclusive evidence that pollutants, namely SO_2, adversely affect coating performance.[8] It was shown that coating performance on prerusted metal substrates was dependent in part upon the time of the year in which the panels were exposed for rusting. A more rapid failure was observed with coatings applied to the rusted bare metal panels recalled during the month of December (higher SO_2 level) as opposed to June (lower SO_2 level). The experiments showed good correlation with the amount of ferrous sulphate formed on the bare metal surface and the subsequent performance of the coating system.

The above cited examples involving primarily actual field exposures suggest that pollutants may indeed exert an experimentally detectable adverse effect on coating performance. Fundamental studies involving the interaction of specific pollutants at controlled, realistic levels with coatings should provide the requisite knowledge to resolve the uncertainty regarding the role of pollutants as a degradative parameter.

II. Project Description

A. Objective

The purpose of this experimental program was to develop laboratory test methods capable of detecting the initial degradation of selected exterior coatings exposed to various pollutant containing environments. Ultimately, the techniques developed in this study will be employed by the Environmental Protection Agency in their laboratory facilities to generate the required additional dose-response and service life data to assess the economic loss attributable to pollutant damage of exterior coatings.

B. Scope of Research

This experimental study involved accomplishing three tasks:

(1) Review of existing exterior exposure records to establish the service life of selected classes of commercially important coatings exposed in a number of locations varying in the general type and level of pollution.

(2) Short term exposure of the selected coatings at four exterior locations (both north and south exposure) which differ in the general type and level of pollution. Attempts will be made to correlate the detectable initial damage from "short-term" outdoor exposure with existing service life data collected in Task I and with the accelerated laboratory results generated in Task III.

(3) Simulate the detectable initial film damage under more controlled, but accelerated weathering conditons in laboratory exposure studies.

III. Materials and Experimental Procedure

A. Coatings

The five coating systems selected for examination in the above three tasks are listed under the following four commercially important paint classes.

1. House Paints

   Lead/titanium/zinc extender in oil with 100% rutile TiO_2.
   Titanium/extender in acrylic latex with 100% rutile TiO_2.

2. Industrial Maintenance Coatings

   Titanium in alkyd with 100% rutile TiO_2.

(3) Coil Coating Finishes

Titanium/extender in urea-alkyd with 75% rutile and 25% anatase TiO_2.

(4) Automotive Refinish

Titanium in nitrocellulose/acrylic with 100% rutile TiO_2.

Although the finishes selected for the industrial maintenance, coil coating and automotive areas are considered somewhat less durable than others in these classifications, they, nevertheless, possess the following desirable characteristics for a study of this nature:

(1) They represent large volume products and, therefore, are considered commercially important.

(2) Extensive histories exist for at least four different geographical locations which differ in environmental conditions.

(3) A considerable amount of performance data versus exterior exposure time has been collected on these coatings over the years.

(4) The relatively short time element for completing Phases II and III of the project necessitated selecting less durable coatings which would be more susceptible to attack by pollutant containing environments.

B. Test Methods for Determining Film Damage

1. Gravimetric Erosion Studies

This method of testing was employed as the main criterion for assessing the performance of selected coatings except for the automotive refinish exposed in Tasks II and III. It is our contention that coatings classified as commercially important have been tested and reformulated sufficiently to avoid the relatively undesirable types of failure such as cracking, delamination, etc. and therefore, "fail" by the relatively desirable mechanism of gradual erosion. In addition, our considerable experience with erosion studies shows that the exterior and Weather-Ometer erosion rates of the selected coatings are normally linear after 3 months and 500 hours of exposure respectively. Consequently, early erosion rates can be used effectively to predict long term durability as well as correlating the results of Task II with those generated in Task III.

For erosion measurements, the weight loss or gain by the control panels maintained in the controlled environment room (77 ± 1° F and 45 ± 2% RH)

was used to adjust the weight loss of the respective exposed coatings at each examination period. This corrected weight determined to the nearest 0.0001 gram was then converted to "dry volume" through a calculated density constant for each coating. The latter value divided by the measured area of coating for each panel gives the film thickness lost in mils resulting from exposure.

The success in generating accurate erosion data is dependent on the periodic removal of accumulated chalk from the coated panel surface.[10,11] During each examination period, the respective coated metal panels were washed using a cotton swab saturated with distilled water containing 5% Tide prior to equilibration and weighing to the nearest 0.001 gram in the controlled environment room.

(2) Infrared Analysis

Attenuated Total Reflectance (ATR) measurements were made on films using a Perkins Elmer Model 621 infrared spectrophotometer equipped with a Wilke Model 12 double beam internal reflection attachment. The ATR reflection process allows penetration of the infrared beam into the sample at a depth approximately equal to one third of the analytical wavelength.

The absorption bands of particular interest in this phase of the study included: (1) The C-H region near 2.9 μ (3400 cm^{-1}), (2) The carboxyl region near 5.8 μ (1740 cm^{-1}), (3) The extender region near 9 μ (1100 cm^{-1}) and (4) The TiO_2 region near 14 μ (730 cm^{-1}). The penetration into the film at these four points would be 1.0, 1.5, 3 and 5 microns, respectively. Care was exercised to achieve optimum contact between the coating and the ATR plate in order to maximize spectra resolution.

(3) Gloss and Sheen Measurements

Gloss [60°] and sheen [85°] measurements were made on all coatings using a Gardner Multi-Angle Glossmeter. All panels were washed with distilled water containing 5% Tide prior to the measurements. These properties are, however, considered of primary importance only for the automotive finish since in the latter case, loss of gloss rather than erosion is the normal mode of failure.

(4) Surface Roughness Measurements

A Talysurf 4 instrument was used to determine the change in topography (micro inch scale) of the control specimens and the five coatings at various exposure conditions. These measurements were performed on panels which had previously received the standard washing procedure.

(5) Instron Determinations

In prior work by the Paint Research Department,[12] tensile strength versus exposure time has been shown to correlate well with the known exterior durability of three of the five selected coatings. The automotive refinish and coil coating are relatively brittle and, therefore, present problems in obtaining reproducible tensile strength results.

The films designated for analysis by stress-strain were sized to 3/4" x 3" with a Precision Sample Cutter*, amalgamated by floating the test sample film side up in a bath of mercury and then conditioned in a controlled environment room for [illegible] hours before testing with an Instron Mechanical Testing Machine.**

* Produced by Tinius Albert Instrument Company

** Produced by Instron Engineering Corporation

Mention of any instrument by product name does not indicate endorsement by the Environmental Protection Agency.

All coatings, excluding the automotive refinish and the latex paint, were tested at a strain rate of 10"/minute. The fragile automotive refinish required a lower test rate of 1"/minute. The latex films, with their lower modulus, were examined at a rate of 100"/minute.

(6) Scanning Electron Microscope Analysis

All five coatings were examined with a Scanning Electron Microscope after selected exposure in either Task II or III to provide a visual record of the topography changes which might be attributable to a given test location or to the aggressive environment in the laboratory exposure study. Two levels of magnification (500X and 1000X) were employed to fascilitate comparison of the physical change that occurred at the coating surface during the exposure condition.

(7) Sorption-Desorption Measurements

Moisture sorption/desorption measurements were incorporated to determine the effect of pollutants on the performance of coatings exposed to laboratory environments. This technique, a simple gravimetric test, has been recently used to evaluate film performance.

The test assembly consists of a four place analytical balance mounted on top of a Plexiglas chamber maintained at constant relative humidity (92 ± 1%) and temperature (77 ± 1° F). A long rod replacing the pan of the balance extended into the chamber so that weighings could be performed without removing the coated panels from the constant sorption atmosphere. An Aminco air unit appropriately attached to the test chamber for proper air flow was used to maintain the desired relative humidity. The entire test assembly was located in a controlled environment room (70° F and 50% RH).

The experiments were performed on the same coated stainless panels used for the erosion rate study. All of the panels possessing an equal area of coating were weighed periodically in the chamber during the 24-hour sorption phase, followed by similar weighings at controlled room conditions for a 24 hour desorption period. This time period was sufficient for the coating to achieve equilibrium constant weight in respect to the specified sorption or desorption conditions. The weight gain or loss (milligrams) by the coatings was standardized by dividing with the respective film thickness previously calculated in the erosion study.

C. The Review of Existing Exterior Exposure Records, Task I

Visual erosion ratings and experimental erosion rate data were used in this phase of study as the main criteria for assessing the service life in years of the five coatings excluding the automotive refinish exposed in at least four of our multiple exterior testing locations. See Table I.

A visual erosion rating of 7, based on a scale of 0 (complete failure) to 10 (no failure), was considered as the minimum value that a coating can reach and still be acceptable in appearance. A coating with a rating below 7 would not have sufficient film thickness to prevent substrate show-through.

In the case of the automotive refinish, the normal mode of failure is not erosion, but rather loss of gloss. Therefore, panels of automotive refinishes are seldom left on exposure for sufficient time (> 5 years) to exhibit visible evidence of erosion. For this finish, the service life in years is equated to the point in time in which the gloss after washing reached 40.

The service life results for some of the coatings were calculated from experimental erosion rate data (mils loss of coating versus exposure time in months). For this conversion, a special set of laboratory panels sprayed with the respective coatings at varying film thicknesses was prepared to establish a graph of visual erosion ratings (9, 8, 7, etc.) versus film thickness. From the latter plots, the film thickness required to give a visual erosion rating of 7 for a given coating was determined. The service life of a coating was then established by entering the difference between the film thickness at its recommended spreading rate and the film thickness equivalent to a visual erosion rating of 7 (from the above plot) in a second graph of experimental film thickness versus exposure time in months.

D. Panel Preparation for Field and Laboratory Exposures

Stainless steel panels (Type [illegible] with a 2B finish), measuring 6 inches by 10.5 inches for the field exposure study and 3 inches by 7 inches for the laboratory exposure study, minimize the effect of erosion on the test results. Five 3/8 inch holes were drilled in each field panel (one at each corner plus one centered along an edge for weighing purposes) for subsequent mounting on 1 foot by 3 foot plywood panels. Only one hole was drilled at the top of each laboratory panel for weighing purposes. The metal panels were then appropriately coded for reference, cleaned in a perchloroethylene vapor degreasing chamber and weighed to the nearest 0.0001 gram with an analytical balance. A taping operation was subsequently performed to confine the area of coating to 6 inches by 9.5 inches for the field panels and 3 inches by 6 inches for the laboratory panels. Care was exercised during handling of the panel to avoid contamination of the surface before the coating operation. The five coatings were then applied directly to the respective panels at controlled uniform film thickness according to the recommended spreading rates (See Table 2.) with a Spraymation automatic spraying unit. Preliminary Weather-Ometer exposure studies showed that all of the coatings would maintain satisfactory adhesion to the unprimed substrates during extended exposure.

TABLE I

Location of Exterior Test Fences

Valparaiso, Indiana*	Miami, Florida*
Detroit, Michigan	New Orleans, Louisiana*
Cleveland, Ohio	Coffeyville, Kansas*
Dayton, Ohio	Los Angeles, California
Gibbsboro, New Jersey	Oakland, California
Atlantic City, New Jersey	Garland, Texas

Montreal, Canada

* Major test sites

TABLE

Application Parameters for the Five Selected Coatings

	Spreading Rate (ft^2/gal.)	Desired Film (Mils)	Mils Achieved*
Automotive Refinish	300	1.5	1.5 ± .1
Latex Coating	300	1.5	1.4 ± .1
Coil Coating	700	1.0	1.1 ± .1
Industrial Maintenance	400	2.1	2.0 ± .1
Oil House Paint	300	2.5	2.6 ± .1

* Averages of 25 panels for each coating

Coatings designated specifically for tensile strength determinations were prepared with a threaded drawdown bar to achieve a minimum 2 mil dry film thickness on tin foil over glass plates. Prior to characterization, the films were removed from their tin foil substrates by amalgamation. This procedure as specified in ASTM D-[illegible]70 precludes damage to the coating caused by stripping from a glass plate or contamination when employing release agents.

All coated panels and films were then aged for at least two weeks in a controlled environment room ([illegible] ± 1° F and [illegible] ± 5% Relative Humidity) to provide adequate cure before determining the initial properties. Finally, the coated metal panels were appropriately cleaned and weighed to the nearest 0.0001 gram. Panels for field exposure including the respective films taped to aluminum panels were mounted with plastic bolts and spacers on coded plywood panels for distribution to the designated exterior testing sites.

E. Exposure Studies

The exposure of the five coatings to field (Task II) and laboratory environments (Task III) enabled the selection of valid test methods for assessing loss in film performance. The successful development of a technique is dependent on generating consistent trends in data for the coatings exposed in both the field and laboratory studies. If correlation in trends exists, then exposure studies employing controlled laboratory environments accompanied by suitable techniques can be used effectively in projecting the economic loss attributable to pollutant damage of coatings exposed in exterior conditions.

1. Field Exposure Study (Task II)

Four exterior locations known to possess different types of pollutants were selected as exposure sites for the five coatings. The location of the sites and the dominant pollutant levels are shown below:

Location	Environment	Pollution Level 1970 Annual Average SO_2 g/M^3	Oxidant μ g/M^3
North Central, North Dakota	clean, rural site	low	low
Los Angeles, California	high oxidant	low	[illegible]4
Chicago, Illinois	high sulfur dioxide	[illegible]7	low
Valparaiso, Indiana	moderate sulfur dioxide	[illegible]2	low

Although the exterior test sites for this portion of the study have been selected to represent a "clean" rural environment, a high SO_2 environment, and a high oxidant environment plus a moderate SO_2 environment, it is recognized that these sites also vary in temperature, relative humidity and ultraviolet light radiation. Since the latter three factors interact simultaneously with the coating, it may

be difficult to discern the effect of pollutant type or level on service life. without elaborate monitoring and data collection instrumentation. Apparat of this nature was considered beyond the scope of the study. However, in spite of this apparent difficulty, the results of each test method can be examined for unusual effects that might be attributable to the pollutant conditions within test locations.

The experimental design, which allows for statistical treatment of the raw data, is shown below:

Experimental Design for Field Exposure Studies

Test Method	Number of Panels Required	
Erosion, Gloss, Sheen, Surface Roughness	replicates x 3 exposure periods x 5 paints x 4 locations x 2 types of exposures (north and south)	320
Intrared,* Scanning Electron Microscope** Tensile Strength	3 exposure periods x 5 paints x 4 locations x 2 types of exposure	240
Controls retained in the controlled environment room	2 replicates x 5 paints x 2 substrates	20
	Total	580

*Not to be replicated
**To be done on 10 films only

A total of 580 panels were prepared (both stainless steel and tin foil substrates) for this exposure study. Twenty of the total number of panels were retained in the controlled environment room for the initial tests and as unexposed controls. The remaining 560 were mounted on exterior test fences located at the four selected locations. This latter number of panels includes an extra two sets of coated metal panels and 3 extra sets of films for each exposure location (both north and south). The additional material was incorporated into the experimental design both for extended exposure beyond the one year contract requirements and as a safety factor in case some panels became damaged. Separate sets of panels were used for each exposure period to avoid the delay in exposure that would be incurred during shipping and testing.

Property evaluation was performed at zero, 4 and 7 months of exposure. Two replicates were used for erosion rate determinations because of the emphasis on this technique. Gloss, sheen and surface roughness measurements were also made on the same set of coated metal panels. The remaining three tests were conducted on the appropriate films. After the analyses were completed for a respective exposure period, the panels were returned to their designated test fence for continued exterior exposure.

Laboratory Exposure Study (Task III)

A Xenon light, Dew Cycle Weather Ometer equipped with automatic control and monitoring devices for SO_2, O_3, UV light, humidity and temperature was used to generate data under controlled, but accelerated weathering conditions.

The environmental variables chosen for this phase of study are shown below:

	Pollutant Level (ppm)		
	Clean	Normal	High
Sulfur Dioxide	0	0.1	1.0
Ozone	0	0.1	1.0

The zero pollutant level represents the control (clean air) with the 0.1 ppm of each pollutant type considered representative of the levels frequently reached in polluted cities. The 1.0 ppm level represents a "highly" polluted industrial site.

The experimental design for the laboratory exposure study which allows for statistical treatment of the raw data is shown below:

Test Method	Number of Panels Required	
Erosion, Gloss, Sheen, Surface Roughness	2 replicates x 5 points x 5 conditions x 2 types of exposure (shaded and unshaded)	100
Tensile Strength, Infrared,* Scanning Electron Microscope**	5 points x 2 conditions x 2 types of exposure x 3 analyses (amounts to eight 1 inch x 1 inch films per point)	150
Control retained in the controlled environment room	2 replicates x 5 points x 2 substrates	20
	Total	270

* Initial, 1 ppm SO_2, 1 ppm O_3
** To be done on 10 films only

A total of 270 panels (both coated stainless steel and films mounted on aluminum substrates) were prepared for Task III. Erosion rate determinations were made on two replicates in the case of either 0.1 ppm SO_2 or O_3 exposures (both north or south) and three or four replicates for the zero pollutant, 1.0 ppm SO_2 or O_3 conditions (north or south). Similarly to Task II, the non-destructive gloss, sheen and surface roughness measurements were performed on the same coated metal panels used for the erosion rate determinations. In most instances, triplicate determinations were made for the tensile strength studies. The 20 panels retained in the controlled environment room were used for determining zero hour (initial) tests and as unexposed controls.

The basic Weather-Ometer schedule selected for all five of the environmental conditions involved one hour of xenon light at about 70% relative humidity and 140° F black panel temperature, followed by one hour of darkness during which dew was condensed on the coated panel or film surface (100% relative humidity and 120° F in the chamber). In addition, one-half of the specimens were shaded from UV light using each environmental condition by the use of a specially designed stainless steel shield.

The Weather-Ometer was calibrated each day of the week to insure that the specified conditions (pollution levels, relative humidity, temperature, etc.) were being properly maintained in the chamber. Continuous monitoring of the pollutant level and the light intensity maintained constant at 340 nanometers with a special filter was provided by strip chart recorder.

Property characterization was performed on the appropriate panels and films at zero, 500, 750 and 1000 hours for each environmental condition. The time required for washing, equilibration and property analyses necessitated intermittent rotation from one set of panels to another to insure that all panels eventually received 1000 hours of exposure to the designated pollutant condition during the one year contract period.

III. Experimental Results and Discussion

A. Task I - The Success of Existing Exterior Exposure Records

1. Introduction

As discussed previously under the Methods and Experimental Procedure section, both visual erosion ratings and gravimetric erosion rate data were used to assess the service life of the selected coatings (excluding the automotive refinish). It was recognized at the onset of this phase of study that sufficient exposure data was available in our files to set the service life for each coating exposed in at least four locations, but not necessarily the same sites in all cases. Greater confidence can be placed on the service life estimates determined by the gravimetric erosion rate studies compared to the visual erosion rate method since many of the variables normally involved in exterior exposure programs and the subjectivity of visual ratings are eliminated.

2. Results and Discussion

The service life data for the five coatings exposed in at least four different exterior locations (except for the roof coating) are presented in Tables [illegible] through [illegible]. Comparison of the overall average service life in years between coatings indicates that the service life house paint is the most durable ([illegible] years - visual ratings), the off-white paint is second in durability ([illegible] years - visual ratings), the roof coating is third in durability (4.95 years - corrective erosion rate data), the alkyd industrial maintenance coating is unexpectedly fourth in durability ([illegible] years - visual ratings), and the automotive refinish system is the lowest in durability ([illegible] years - based on 20 gloss after washing of [illegible]). It is also noted that within each of the five

paint systems the range in average years required to reach an erosion rating of seven or a gloss after washing of 40 is approximately two to three years.

Further examination of Tables 3 through 7 shows that the width of ranges in months required to reach an erosion rating of seven or a gloss after washing of 40 is fairly large for the two house paints and the alkyd industrial paint exposed in Valparaiso, Indiana and for the automobile coating exposed in Miami, Florida. However, the magnitude of the ranges is not entirely unreasonable when considering all of the potential variables entering into the visual erosion rating (influence of substrate type, primer differences, film thickness variations, different evaluators, etc.)

TABLE 3

Service Life for Acrylic Latex House Paint Exposed at Five Exterior Exposure Sites

Location Vertical South	Ave. Yrs to Erosion Rating of 7	Ave. Mos. to Erosion Rating of 7	Range in Mos.	No. of Panels
Valparaiso, Ind.	7.75	93	60-136	20
Miami, Fla.	6.17	74	54-85	6
Wilmington, Del.	5.16	62	48-66	6
Newton, Pa.	5.08	61	--	1
Palmerton, Pa.	7.92	95	70-107	3
Overall Average	6.41			

TABLE 4

Service Life Data for L/T/Z Oil Base House Paint Exposed at Five Exterior Expos. Sites

Location Vertical South	Ave. Yrs. to Erosion Rating of 7	Ave. Mos. to Erosion Rating of 7	Range in Mos.	No. of Panels
Valparaiso, Ind.	5.83	70	44-118	29
Miami, Fla.	4.00	48	28-60	9
Wilmington, Del.	4.17	50	45-60	5
Concord, Calif.	5.42	65	63-66	2
Palmerton, Pa.	6.5	78	--	1
Overall Average	5.18			

TABLE 5

Service Life Data for the Alkyd Industrial Maintenance Coating Exposed at Five Exterior Exposure Sites

Location Vertical South	Ave. Yrs. to Erosion Rating of 7	Ave. Mos. to Erosion Rating of 7	Range in Mos.	No. of Panels
Valparaiso, Ind.	4.58	55	0	2
Miami, Fla.	4.50	54	42-60	5
Oakland, Calif.	4.16	50	-	1
Atlantic City, N.J.	3.25	39	-	1
Garland, Texas	5.75	69	-	1
Overall Average	4.28			

TABLE 6

Service Life Data for Nitrocellulose Acrylic Automobile Finish Exposed at Four Exterior Exposure Sites

Location 45° South	Ave. Yrs. to 40 Gloss	Ave. Mos. to 40 Gloss	Range in Months	Number of Panels
Valparaiso, Ind.	2.33	28	-	1
Miami, Florida	5.67	68	13-65	58
Wilmington, Delaware	2.42	29	22-36	5
Memphis, Tenn.	1.83	22	-	1
Overall Average	2.29			

TABLE 7

Service Life in Years from Erosion Rate Data versus Visual Ratings for the Coil Coating, Acrylic Latex and Oil Paint

Paint	Erosion Rate Data* Valparaiso	Miami	Visual Ratings** Valparaiso	Miami
Coil Coating (45° South)	5.42 yr. (Unprimed) (Overall average 4.50)	4.51 yr. (Unprimed)	-- --	-- --
Acrylic Latex (85° South)	5.90 yr. (Unprimed) 5.70 (Dark primer)	-	7.75 yr.	6.17 yr.
L/T/Z Oil Base Paint (85° South)	8.94 yr. (Unprimed 7.9 yr. (Dark primer)		5.83 yr.	4.00 yr.

* Determined from experimental erosion rate data (mils loss versus exposure time)
** Previous data presented in Tables 3 and 4

B. Task II - Short Term Exterior Exposure

(1) Introduction

Only two data points (three and seven months for each test method were collected during Task II. Consequently, the limited data essentially precludes statistical treatment and restricts only the reporting of apparent trends which may conceivably change consequent to additional exterior exposure.

(2) Results and Discussion

(a) Gravimetric Erosion Studies

The discussion of erosion studies for Task II will be centered on the 7 months results since the latter time period is nearly equivalent to the point where the erosion rate becomes linear. Consequently, greater confidence can be placed on the existing trends in data.

Examination of the data presented in Table 2 shows the expected consistency in the level of erosion (mils lost) being greater particularly for the exposed alkyd industrial soft oil based paint panels (south location) compared to the specimens on the unexposed or north side of the test fence. It is also apparent, particularly for the exposed panels, that the mils loss of coating is higher at Chicago and Valparaiso than at the Los Angeles and North Dakota locations. These results may be explained by the higher SO_2 levels at the former two exposure sites. As will be revealed during the discussion in Task III, this trend is consistent with the Weather-Ometer studies which show that SO_2 at concentrations representative of a highly polluted industrial site (1 ppm) produced a greater adverse effect on the level of erosion than O_3 at similar levels for specific coatings. Rainfall, sunlight, UV light intensity, and pollutant type and level were not monitored at the various test sites to confirm the proposed effect of pollutants on exterior erosion levels. It is also significant from the standpoint of correlation that the ranking in terms of mils lost (magnitude of erosion) for the coatings in Table 2 is virtually the same as in the accelerated studies regardless of exposure location (both north and south).

TABLE

Erosion Data Mils Loss of the Selected Coatings after 7 Months Exposure at Four Exterior Locations Both North and South

Coatings	Exposure Site Location: Research Center, North	Research Center, South	Valparaiso, Indiana, North	Valparaiso, Indiana, South	Los Angeles, California, North	Los Angeles, California, South	Leeds, North Dakota, North	Leeds, North Dakota, South
Automotive Refinish	.01	.01	.01	.02	.01	.01	.01	.02
Coil Coating	.02	.03	.03	.04	.04	.05	.01	.02
Latex Coating	.06	.07	.07	.07	.04	.04	.03	.06
Industrial Maintenance	.07	.11	.09	.11	.08	.08	.06	.07
Oil House Paint	.10	.11	.11	.12	.10	.13	.07	.10

b Infrared Analysis

Attenuated Total Reflectance ATR measurements were performed on films of the selected coatings exposed for and months only at the Chicago Research Center location both north and south exposures . Emphasis will be placed on the spectra for the month exposed samples since they were very similar to those for the month exposure except for the expected greater binder degradation. This degradation was even more pronounced for the coatings exposed on the south position of the test fence. The extent of binder breakdown was determined by the reduction in intensity of the ester carbonyl and C-H bands relative to pigment absorption, specifically TiO_2.

The oil and alkyd systems (oil house paint, industrial maintenance, coil coating were degraded to a greater extent than the two acrylic coatings (latex, automotive refinish consequent to the weathering conditions at the Chicago test site. In addition, the former three coatings showed a considerable increase in metallic soap content. Concentration of driers at the surface during weathering or formation of metallic soaps of fatty acids and/or degradation products could account for this observation.

The oil house paint and coil coating which contain a moderate amount of $CaCO_3$ extender exhibited a considerable decrease in carbonate content accompanied by the formation of sulphite and/or sulfate compounds. This formation was detected irrespective of north or south exposure and despite the fact that the samples were washed to remove any accumulated chalk or dirt prior to ATR analysis. The sulfite or sulfate compound could be formed by the reaction of the dissociated inert cation (in this case, Ca^{++}) with sulfurous acid (absorbed $SO_2 + H_2O$) and/or sulfuric acid ($SO_3 + H_2O$) that is present in the paint film.

The results of this phase of study provided the initial evidence that SO_2 at moderate to high concentrations exerts a detectable adverse effect on coatings particularly containing inerts susceptible to dissolution in acidic media. The disapperance of the inert through formation of water soluble compounds would be reflected in high apparent erosion levels which, in part, supports the contention that the higher erosion encountered at Chicago and Valparaiso versus Los Angeles and North Dakota may be attributable to the SO_2 level.

c) Gloss and Sheen Measurements

Gloss (60^o) and Sheen (85^o) measurements of the selected coatings were made after 5 and 7 months exposure at the four test sites (both north and south positions) using a Gardner Multi-Angle Glossmeter. All panels had been washed as a standard procedure and weighed for the erosion studies prior to characterization by gloss and sheen. These methods of testing are, however, considered of primary importance particularly for the automobile refinish since in the latter case, loss of gloss rather than erosion is the normal mode of failure. The service life for the latter coating is equated to the point in time in which the gloss after washing reaches 40.

Gloss measurements provided more consistent trends than sheen particularly for the automotive refinish and the industrial maintenance coating which exhibit a high initial gloss and/or a gradual decrease in gloss during exposure. As shown in Table 9, the gloss for the former two coatings is higher (as expected) for the north side position at all four locations. It is also noted that the automotive refinish is retaining a relatively high gloss level. In addition, the level of gloss for the industrial maintenance coating is lower at Valparaiso and Chicago versus Los Angeles and North Dakota. The latter trend suggesting greatercoating degradation is consistent with the erosion and ATR results previously presented. The other three coatings (oil, latex and coil) exhibit a relatively low gloss irrespective of exposure location.

The results of the sheen determinations presented in Table 10 do not at this time correlate with the trends discernible from the gloss data after 7 months exposure. However, detectable differences specific to a test site may develop with additional exposure since the sheen of four coatings is presently at a reasonable level.

TABLE 3

Gloss Measurements (60°) of the Selected Coatings after 3 and 7 months of Exposure at the Four Exterior Locations

Exterior Test Site Location

Valparaiso, Indiana

	Initial	North 3 months	North 7 months	South 3 months	South 7 months
Automotive Refinish	92	92	92	92	91
Industrial Maintenance	57	30	30	30	22
Oil House Paint	12	6	5	10	5
Coil Coating	5	4	5	4	3
Latex Coating	5	5	5	5	5

Research Center, Chicago

	Initial	North 3 months	North 7 months	South 3 months	South 7 months
Automotive Refinish	89	89	89	87	87
Industrial Maintenance	57	57	25	31	20
Oil House Paint	12	9	5	7	4
Latex Coating	5	5	5	5	5
Coil Coating	5	5	4	5	3

Los Angeles, California

	Initial	North 3 months	North 7 months	South 3 months	South 7 months
Automotive Refinish	89	92	90	85	84
Industrial Maintenance	57	32	30	35	25
Oil House Paint	12	8	5	5	5
Latex Coating	5	5	5	5	5
Coil Coating	5	4	4	4	4

Leeds, North Dakota

	Initial	North 3 months	North 7 months	South 3 months	South 7 months
Automotive Refinish	89	89	88	87	86
Industrial Maintenance	57	33	32	26	24
Oil House Paint	12	12	5	10	4
Latex Coating	5	5	5	5	5
Coil Coating	5	5	5	5	5

TABLE 10

Sheen Measurements[1] of the Selected Coatings After 7 and 7 Months Exposure at the Four Exterior Locations

Coatings	Initial	7 months	7 months	7 months	7 months
	Exterior Test Site Location				
	Valparaiso, Indiana				
		North		South	
Automotive Refinish					74
Industrial Maintenance					55
Oil House Paint	1		1		1
Coil Coating	1	11	11	11	11
Latex Coating					3
	Research Center, Chicago				
		North		South	
Automotive Refinish					72
Industrial Maintenance					40
Oil House Paint	1	1	1	1	1
Coil Coating	1	10	10	10	10
Latex Coating					3
	Los Angeles, California				
		North		South	
Automotive Refinish					70
Industrial Maintenance					40
Oil House Paint	1				10
Coil Coating	1	1	10	10	10
Latex Coating					3
	Leeds, North Dakota				
		North		South	
Automotive Refinish					74
Industrial Maintenance					55
Oil House Paint	1				22
Coil Coating	1	1	1	1	12
Latex Coating					3

d Surface Roughness Measurements

A Talysurf 4 instrument was used to determine the change in topography (microinch scale) of the unexposed controls and the five coatings exposed only for 7 months at the four exterior test sites. These measurements were performed on the erosion panels which had been washed with a 5% Tide solution to minimize the influence of dirt accumulation on the test results.

The data collected on the exposed coatings (both north and south) were converted initially to a percent change in surface roughness (S.R. with respect to the unexposed control values (% change = $\frac{\text{S.R. exposed - S.R. control}}{\text{S. R. control}}$).
A positive value reflects an increase in surface roughness resulting from exposure whereas as a zero or negative percent change indicates no change or decreased surface roughness respectively, compared to the unexposed, control value. Subsequently, the data for a given coating in Table 11 were ranked on a scale from 1 (highest % change in surface roughness) to 5 (lowest % change), both within and between test sites. These data are presented in Table 12. Coatings with equivalent change in surface roughness were given identical rankings. Summation of the individual rankings in columns (within location) provides five numbers for subsequent comparison of the magnitude of change in surface roughness specific to a given coating irrespective of test site. Similarly, summation of the rankings in rows (between location) facilitates establishing the effect of the test site on the level of surface roughness independent of a coating type. The trends in data either within or between test sites are then determined by comparing the magnitude of the summations. A low value indicates a high percentage change in surface roughness whereas a high summation connotes a low change in roughness.

Examination of the results presented in Table 12 for ranking within test site shows that the automotive refinish exhibited the greatest change in surface roughness followed by the oil house paint, industrial maintenance, coil coating and latex paint, respectively. This trend is apparent regardless of panel position (shaded or unshaded) on the test fence. Comparison between locations illustrates that the Valparaiso and Los Angeles test sites effected a higher percentage change in surface roughness for the five coatings than exposure at the Research Center and Leeds, North Dakota locations (both north and south).

Consideration of the results presented for the erosion (Table 8) and surface roughness techniques, shows limited consistency with respect to the ranking in level of property for the five coatings. The inconsistency is particularly dramatic for the automotive refinish which exhibited the highest percentage change in surface roughness but yet high gloss retention and the lowest level of ersoion.

It is apparent, however, that agreement in trends exists from the standpoint of extremes in exposure location. The Valparaiso test site produced the greatest change in both erosion and surface roughness whereas Leeds, North Dakota showed the least effect.

TABLE II

Surface Roughness: Percent Change with Respect to Unexposed Control of the Selected Coatings After 3 Months Exposure

Coatings	Exposure Test Site Location	
	Valparaiso, Indiana	
	North	South
Automotive Refinish	[illegible]	[illegible]
Oil House Paint	[illegible]	[illegible]
Industrial Maintenance	[illegible]	[illegible]
Coil Coating	[illegible]	[illegible]
Latex Paint	[illegible]	1.1
	Research Center, Chicago	
	North	South
Automotive Refinish	[illegible]	[illegible]
Oil House Paint	[illegible]	[illegible]
Industrial Maintenance	[illegible]	[illegible]
Coil Coating	[illegible]	[illegible]
Latex Paint	[illegible]	[illegible]
	Los Angeles, California	
	North	South
Automotive Refinish	[illegible]	[illegible]
Oil House Paint	[illegible]	[illegible]
Industrial Maintenance	[illegible]	[illegible]
Coil Coating	[illegible]	[illegible]
Latex Paint	[illegible]	1.1
	Leeds, North Dakota	
	North	South
Automotive Refinish	[illegible]	[illegible]
Oil House Paint	[illegible]	[illegible]
Industrial Maintenance	[illegible]	[illegible]
Coil Coating	[illegible]	[illegible]
Latex Paint	[illegible]	[illegible]

TABLE 1

Ranking of Percent Change in Surface Roughness for the Five Selected Coatings Exposed for 7 Months at the Four Exterior Locations

COATINGS

Exposure Test Site Location	Automotive Refinish	Latex Coating	Coil Coating	Industrial Maintenance	Oil House Paint
Ranking Within Location - Shaded					
Valparaiso, Ind.	1	3	4	5	2
Research Cen., Chgo.	1	4	5	3	2
Los Angeles, Calif.	1	3	5	4	2
Leeds, North Dakota	1	4.5	4.5	3	2
Summation	4	14.5	18.5	15	8
Ranking Within Location - Unshaded					
Valparaiso, Ind.	1	4	5	3	2
Research Cen., Chgo.	1	5	3	4	2
Los Angeles, Calif.	1	5	3	4	2
Leeds, North Dakota	1	5	4	3	2
Summation	4	19	15	14	8
Ranking Between Location - Shaded					
Valparaiso, Ind.	3	1	1.5	1	2 = 7.5
Research Cen., Chgo.	2.5	2.5	5.5	2	3 = 14.5
Los Angeles, Calif.	1	2.5	1.5	3	1 = 9.0
Leeds, North Dakota	3.5	4	3.5	4	4 = 19
Ranking Between Location - Unshaded					
Valparaiso, Ind.	1	2	3.5	1	2 = 9.5
Research Cen., Chgo.	3	1	2	2.5	3 = 11.5
Los Angeles, Calif.	3	3	1	2.5	1 = 10.5
Leeds, North Dakota	2	4	3.5	4	4 = 18.5

e. Stress-Strain Determinations

Both tensile strength and elongation were determined during the analysis of free films with the Instron Testing Machine. Major emphasis is placed on the property of tensile strength since it exhibits an inversion or inflection point with exposure time indicating that the process of chain-scissioning commences to predominate over crosslinking. Elongation is a less desirable property due to the gradual exponential decrease during weathering. Coatings with good to excellent durability typically show a gradual positive slope before and a negative slope after the inversion point in tensile strength. Conversely, poorer performing coatings exhibit a rather abrupt transition in tensile strength versus exposure time. Consequently, this approach to property characterization can be used to assess the service life of a wide variety of coatings.[2] Relatively brittle coatings represent an exception since they tend to give non-reproducible tensile strength results.

Examination of the tensile strength results for the latex, oil and alkyd industrial maintenance coatings presented in Table I show an expected increase in tensile strength with exposure time for both the north and south positions. The brittle automotive refinish and coil coatings are exhibiting erratic tensile strength behavior for all exposure sites. Additional exposure time will be required for generation of sufficient tensile strength data to establish the effect of test location and to provide service life projections.

f. Scanning Electron Photomicrographs

The coatings exposed for 7 months on the south position only of the Research Center and Leeds, North Dakota test sites were selected for examination by scanning electron microscopy. As previously noted these locations represent the extremes in the level of SO_2 and produced the greatest difference in the results generated by the erosion study. Unexposed controls were also included in the series for comparison.

Figures 1 through 21 are arrarged according to a coating type depicting the unexposed control, Leeds, North Dakota and Research Center photomicrographs for the automotive refinish, latex house paint, coil coating, alkyd industrial maintenance coating and oil house paint, respectively. It is readily apparent that the Research Center location caused a greater topographical change for the coil and industrial maintenance coating than exposure to Leeds, North Dakota. The automotive refinish appears unaffected by exposure to either location. Examination of the latex paint at 7000X (Figures 6 and 8) shows essentially no difference in the amount of surface degradation between the two exposure sites. These observations are generally consistent with the trends in results reported for the erosion studies and the surface roughness measurements. An apparent deviation specifically with the erosion data (Table 3) exists for the oil house paint. As shown in Figures 19 and 21 (7000X), a greater quantity of exposed pigment suggesting more extensive binder degradation is evident on the Leeds, North Dakota samples despite the fact that the Research Center location produced the highest level of erosion.

TABLE I*

Tensile Strength Determinations (PSI) of the Selected Coatings after 5 and 7 Months Exposure at Four Exterior Locations Both North and South

Coatings	Initial	Exterior Test Site Location Valparaiso, Indiana North 5 months	North 7 months	South 5 months	South 7 months
Automotive Refinish	[illegible]	[illegible]	100	[illegible]	1050
Coil Coating	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
Latex Coating	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
Industrial Maintenance	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
Oil House Paint	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]

	Initial	Research Center, Chicago North 5 months	North 7 months	South 5 months	South 7 months
Automotive Refinish	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
Coil Coating	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
Latex Coating	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
Industrial Maintenance	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
Oil House Paint	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]

	Initial	Los Angeles, California North 5 months	North 7 months	South 5 months	South 7 months
Automotive Refinish	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
Coil Coating	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
Latex Coating	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
Industrial Maintenance	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
Oil House Paint	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]

	Initial	Leeds, North Dakota North 5 months	North 7 months	South 5 months	South 7 months
Automotive Refinish	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
Coil Coating	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
Latex Coating	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
Industrial Maintenance	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
Oil House Paint	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]

* Tensile strength rounded to nearest 50 PSI

AUTOMOTIVE REFINISH

Figure 1 - Unexposed Control 700X

Figure 2 - 7 Months at Leeds, North Dakota 700X

Figure 3 - " " " Research Center, Chicago 7000X

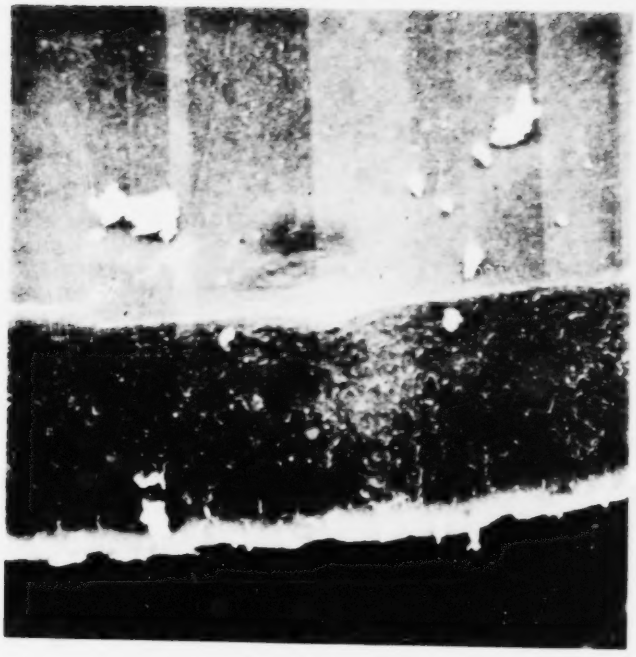


Fig. 1

Fig. 2

Fig. 3

LATEX HOUSE PAINT

Figure 4 - Unexposed Control	700X
Figure 5 - 7 Months at Leeds, North Dakota	700X
Figure 6 - " " " " " "	7000X
Figure 7 - " " " Research Center, Chicago	700X
Figure 8 - " " " " " "	7000X

Fig. 4

Fig. 5

Fig. 6

Fig. 7

Fig. 8

COIL COATING

Figure 9 - Unexposed Control 700X

Figure 10 - 7 Months at Leeds, North Dakota 700X

Figure 11 - " " " Research Center, Chicago 7000X

Fig. 9

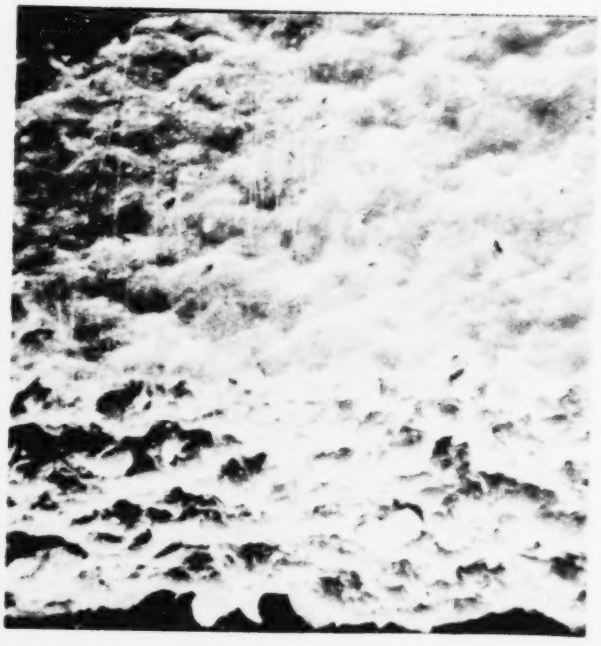


Fig. 10

Fig. 11

INDUSTRIAL MAINTENANCE COATINGS

Figure 12 - Unexposed Control 700X

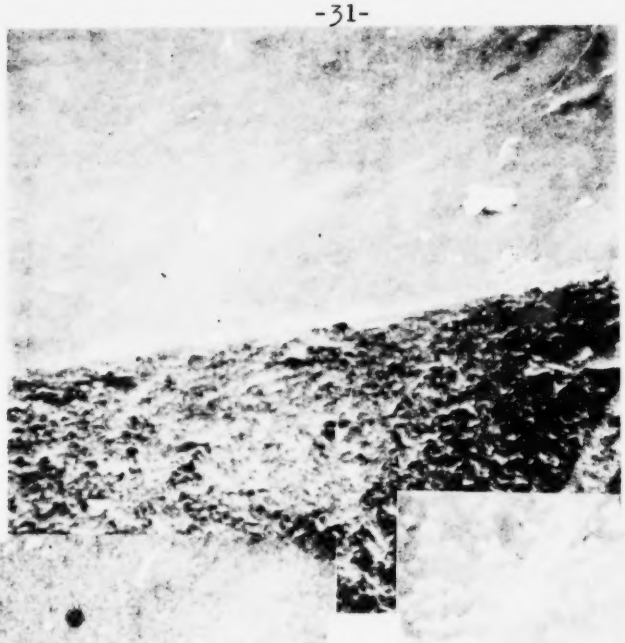
Figure 13 - 7 Months at Leeds, North Dakota 700X

Figure 14 - " " " " " " 7000X

Figure 15 - " " " Research Center, Chicago 700X

Figure 16 - " " " " " " 7000X

Fig. 12



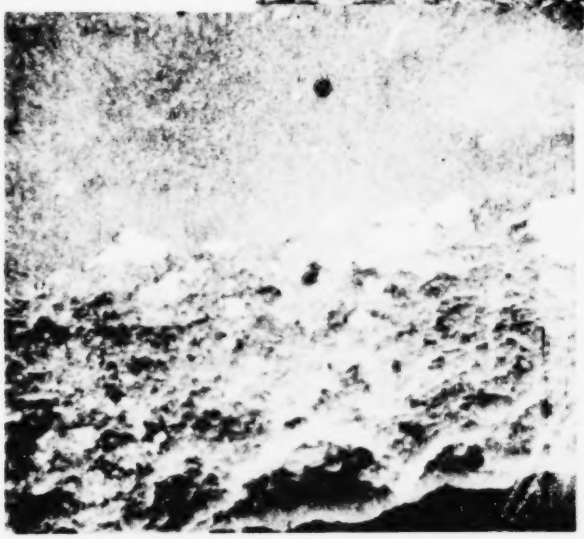


Fig. 13

Fig. 14

Fig. 15

Fig. 16

OIL HOUSE PAINT

Figure 17 - Unexposed Control 700X

Figure 18 - 7 Months at Leeds, North Dakota 700X

Figure 19 - " " " " " " 7000X

Figure 20 - " " " Research Center, Chicago 700X

Figure 21 - " " " " " " 7000X

Fig. 17

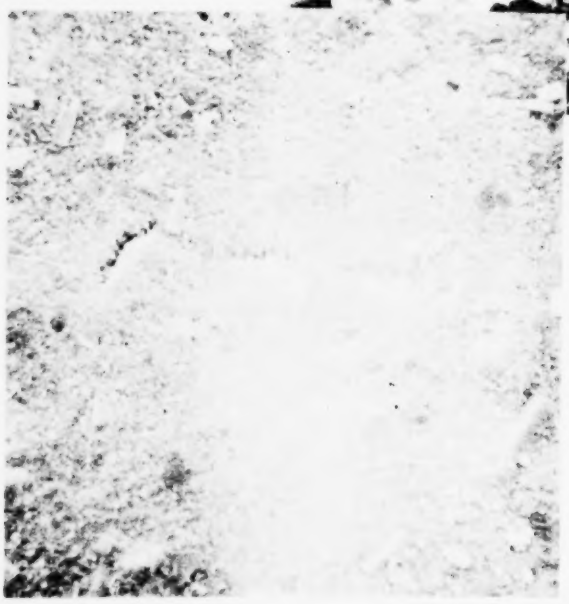


Fig. 18

Fig. 19

Fig. 20

Fig. 21

C. Task III - Accelerated Laboratory Exposures

1. Introduction

This phase of the study provided the most meaningful results due to the self-contained experiments permitted with Weather-Ometer exposures coupled with accurately controlled environments and frequent examination periods. It is recognized, however, that the high relative humidity (70-100%) that was maintained in the Weather-Ometer cabinet could conceivably cause higher rates of degradation of specific coatings, particularly when SO_2 is present. In this latter case, sulfurous acid formed from the reaction of SO_2 and water on or within the film will continuously be in contact with the coating because the concentration of SO_2 was maintained essentially constant at 0.1 or 1.0 ppm during the entire exposure period. The latter condition may be too accelerated compared to exterior exposure where the coatings are intermittently subjected to a relatively low level of sulfurous acid. It is possible, however, that the above contention could be premature since the pH of the water droplets formed on the panel surface during the dew cycle and those present during the initial portion of the 'light on' phase was 3 for both the shaded and unshaded condition (0.1 and 1.0 ppm SO_2 exposures). Also the dew on the panels located at the Research Center test site showed a pH of 3. The limited time for this study did not permit a repeat experiment to determine whether the detectable effects of pollutants such as SO_2 on coatings properties is still real under identical environmental conditions except for a lower relative humidity during the light-on phase of the cycle.

2. Results and Discussion

(a) Gravimetric Erosion Studies

Linear regression analyses were performed on the erosion data (mils loss) for the five coatings exposed in the Weather-Ometer for 1000 hours to either the zero pollutant, the 0.1 or 1.0 ppm SO_2 and the 0.1 or 1.0 ppm O_3 levels (shaded and unshaded conditions). Only the data points collected at 400, 700 and 1000 hours of exposure were used in determining the slope (rate of erosion) since the curves for most of the coatings (excluding the oil base, shaded condition, 1.0 ppm SO_2 and 1.0 ppm O_3, and unshaded 0.1 ppm O_3 and 1.0 ppm O_3; latex, unshaded, 0.1 ppm SO_2) are linear during this exposure period for both the shaded and unshaded conditions. The zero hour data point was excluded from the analysis because the slope of virtually all erosion rate curves during the zero to 400 hour period is somewhat greater than the latter part (400-1000 hours) due primarily to the removal of water soluble materials. In essence, the contribution of the weight loss or mils loss attributable to the removal of the water sensitive materials is eliminated from consideration. Consequently, meaningful conclusions concerning the erosion due mainly to loss of binder and pigment can be made by simply comparing slopes. The intercept of the curve will be ignored in the discussion of

results since the magnitude of the intercept is dependent on the extent of water soluble loss.

It is apparent from the above discussion that erroneous conclusions may result if based solely on erosion values (mils loss) after a given exposure period. For example, two coatings may show different mils loss but equal rates of erosion after 1000 hours exposure in a Weather-Ometer. Providing the difference in the magnitude of erosion was caused by loss of water soluble materials, both coatings should exhibit equivalent service life within the environment under consideration.

The slopes and 95% confidence limits for the erosion data (mils loss) collected after 500, 750 and 1000 hours exposure are presented in Table 14. It is readily apparent that the confidence limits for the slopes of a number of coatings are relatively wide. This situation is commonly encountered with a limited number of data points. Consequently, T-test probabilities were calculated to facilitate discerning whether there is a statistically significant difference between the slope (rate of erosion) of a given coating exposed to any pollutant type or level versus the slope for the respective zero pollutant exposure. Comparisons can also be made between the effect of pollutant types or levels on a given coating within an exposure condition (shaded or unshaded) because all probabilities were based on the respective zero pollutant level slope.

The criterion of statistical significance is often arbitrarily set at 95 percent confidence. On a practical basis, however, reasonable confidence can be expressed when there is an 80 percent probability that a difference exists between two estimates derived from relatively limited data. Increasing the number of data points would normally narrow the confidence limits for a given statistic providing justification for comparing at a high level of confidence such as 95 percent.

A further clarification should be made concerning statistical versus practical significance. It is noted in Table 14 that there is a 92 percent probability that 1.0 ppm SO_2 (unshaded condition) affected the automotive refinish compared to the zero pollutant level. Despite the statistical significance, the automotive refinish lost only 0.02 mils and 0.05 mils after exposure in the unshaded condition to the zero pollutant and 1.0 ppm SO_2 level respectively. Consequently, there is no practical difference in terms of mils loss for the coating in question. Similar reasoning can also be applied for the automotive refinish regardless of the pollutant type or level in either the shaded or unshaded conditions.

The majority of coatings exposed to 0.1 ppm O_3 and to a lesser extent with 0.1 ppm SO_2 both shaded and unshaded conditions) exhibited a slope (Table 14) lower than the respective zero pollutant slope. This trend in data contradicts the expected behavior of increasing rates of erosion upon exposure to higher levels of pollutant for both SO_2 or O_3. The deviation from the expected trend can be reasonably explained by considering the influence of adsorption of O_3

TABLE 10

Slope of Erosion Data Accompanied by a T-Test Probability (%) that a Statistical Difference Exists Between the Respective Slope for a Given Pollutant Type and Level Versus the Zero Pollutant level

SHADED

Coating	0	SO_2 0.1 ppm	SO_2 1.0 ppm	O_3 0.1 ppm	O_3 1.0 ppm
Oil	[illegible]	[illegible] 91.5%	47.0±10.6 [illegible]%	8.5±[illegible] 25%	10.6± 3.0 50%
Industrial Maintenance	10.[illegible]	[illegible] [illegible]%	12.[illegible] 50%	[illegible].1±[illegible].8 75%	21.1±15 [illegible]%
Coil	[illegible]	[illegible] [illegible]%	1[illegible]±10.5 [illegible]%	0.[illegible]±.1 70%	4.1±4.2 55%
Latex	0.[illegible]±1.7	[illegible].0±[illegible].0 [illegible]%	7.0±[illegible].0 [illegible]%	[illegible].[illegible]±4.4 72%	[illegible].[illegible]±1.0 [illegible]%
Automobile Refinish	1.1±1.[illegible]	1[illegible]±[illegible].[illegible] [illegible]%	[illegible].[illegible]±1.[illegible] [illegible]%	0.[illegible]±[illegible].[illegible] 13%	[illegible].0±1.[illegible] [illegible]%

UNSHADED

Coating	0	SO_2 0.1 ppm	SO_2 1.0 ppm	O_3 0.1 ppm	O_3 1.0 ppm
Oil	20.1±[illegible].[illegible]	[illegible].[illegible].0 [illegible]%	1[illegible].0±[illegible].0 [illegible]%	[illegible]±[illegible].[illegible] [illegible]%	[illegible].[illegible]±10.[illegible] [illegible]%
Industrial Maintenance	[illegible]±[illegible].1 [illegible]%	[illegible].[illegible]±[illegible].[illegible] [illegible]%	[illegible].[illegible]±7.0 [illegible]%	[illegible].[illegible]±[illegible].1 [illegible]%	[illegible].1±14.0 [illegible]%
Coil	11.[illegible]	[illegible].[illegible]±1.7 [illegible]%	[illegible].1±[illegible].7 [illegible]%	[illegible].[illegible]±[illegible].[illegible] [illegible]%	[illegible].[illegible]±[illegible].[illegible] [illegible]%
Latex	[illegible].[illegible]±1.[illegible]	[illegible].[illegible]±[illegible].[illegible] 14%	11.1±1.0 [illegible]%	2.[illegible]±0.[illegible] [illegible]%	[illegible].[illegible]±[illegible].[illegible] [illegible]%
Automotive Refinish	1.8±0.[illegible]	[illegible].[illegible].0 [illegible]%	[illegible].1±2.[illegible] 75%	1.[illegible]±1.7 13%	[illegible].1±1.[illegible] [illegible]%

by the binder on the apparent rate of degradation (erosion). Both of these processes are logically occurring simultaneously during exposure in the Weather-Ometer. The weight gain contribution due to adsorption of pollutant by the binder would ultimately reflect a lower level of erosion. At the 0.1 ppm SO_2 or O_3 levels, there is apparently sufficient adsorption of pollutant to produce a level of erosion lower than the respective zero pollutant condition. In the case of the 1.0 ppm SO_2 or O_3 exposures, the degradation process almost completely predominates compared to adsorption, resulting in significantly higher rates of erosion than the zero pollutant condition. However, it should be re-emphasized that the adsorption phenomenon would be most likely to occur during exposure to O_3 regardless of the pollutant level. This condition would tend to magnify any differences that exist between the level of erosion produced by SO_2 versus O_3.

In view of the above conflicting trend in data, major emphasis during the subsequent discussion will be placed on the comparisons between the rates of erosion for the zero and 1.0 ppm SO_2 or O_3 pollutant levels. Examination of the probabilities for the 1.0 ppm SO_2 pollutant level (within either the shaded or unshaded condition) shows that SO_2 caused a statistically significant effect on the oil house paint, latex paint and the urea/alkyd coil coating, but no effect on the alkyd industrial maintenance and nitrocellulose/acrylic coatings. It is an important point to note that the latter two paints which were unaffected by exposure to SO_2 are essentially free of metallic silicate or calcium carbonate extenders. The other three paints that exhibited significant effects from SO_2 all contain aluminum or magnesium silicate or calcium carbonate or combinations of these inerts.

In contrast to the trend in data for the 1.0 ppm SO_2 exposure, O_3 at the 1.0 ppm level affected all coatings (excluding the nitrocellulose/acrylic) in the unshaded condition. Furthermore, the alkyd industrial maintenance coating was significantly affected by O_3 regardless of exposure in the shaded or unshaded condition. In view of the results for both SO_2 and O_3 exposures, the attack by SO_2 appears to be mainly on the extender component (absent in the industrial maintenance and automotive refinish whereas O_3 almost exclusively attacks the binder through a chain scissioning process. However, because of the general similarity of the oil vehicle with respect to the alkyd, it is not entirely clear why the oil house paint was unaffected by 1.0 ppm O_3 in the shaded condition.

The slopes of the five coatings with accompanying 95 percent confidence levels plotted versus pollutant level (zero, 0.1 ppm SO_2 or O_3, and 1.0 ppm SO_2 or O_3) provide easy comparison of the composite data in a few graphs. The plots presented in Figures 22 through 25 demonstrate that the previously discussed anomalies associated with exposure of the coatings to 0.1 ppm SO_2 or O_3 do not for the most part negate the a priori hypothesis of erosion rates or mils loss being linearly related to pollutant concentration. Consequently, these curves along with similar exterior erosion data collected over an extended exposure period (2 years minimum) can be used as a method for predicting the effect of any level of pollutant on coating durability.

Figure 32. Slopes of Erosion Data Versus Pollutant Level for the Coating Exposed to SO_2 in the Weather-Ometer-Ometer Shaded Condition

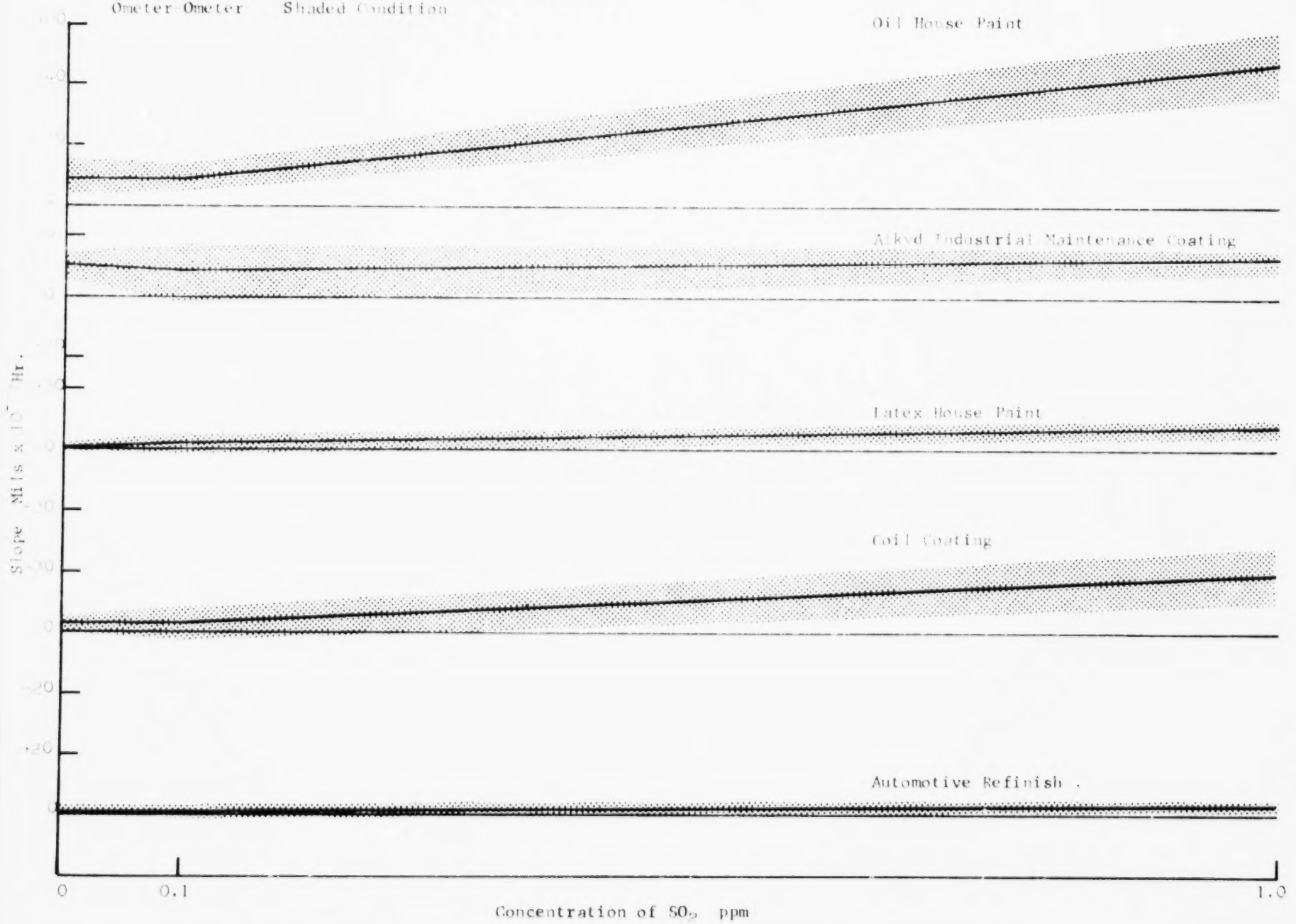


Figure - Slopes of Erosion Data Versus Pollutant Level for the Coating Exposed to SO_2 in the Weather Ometer Unshaded Condition

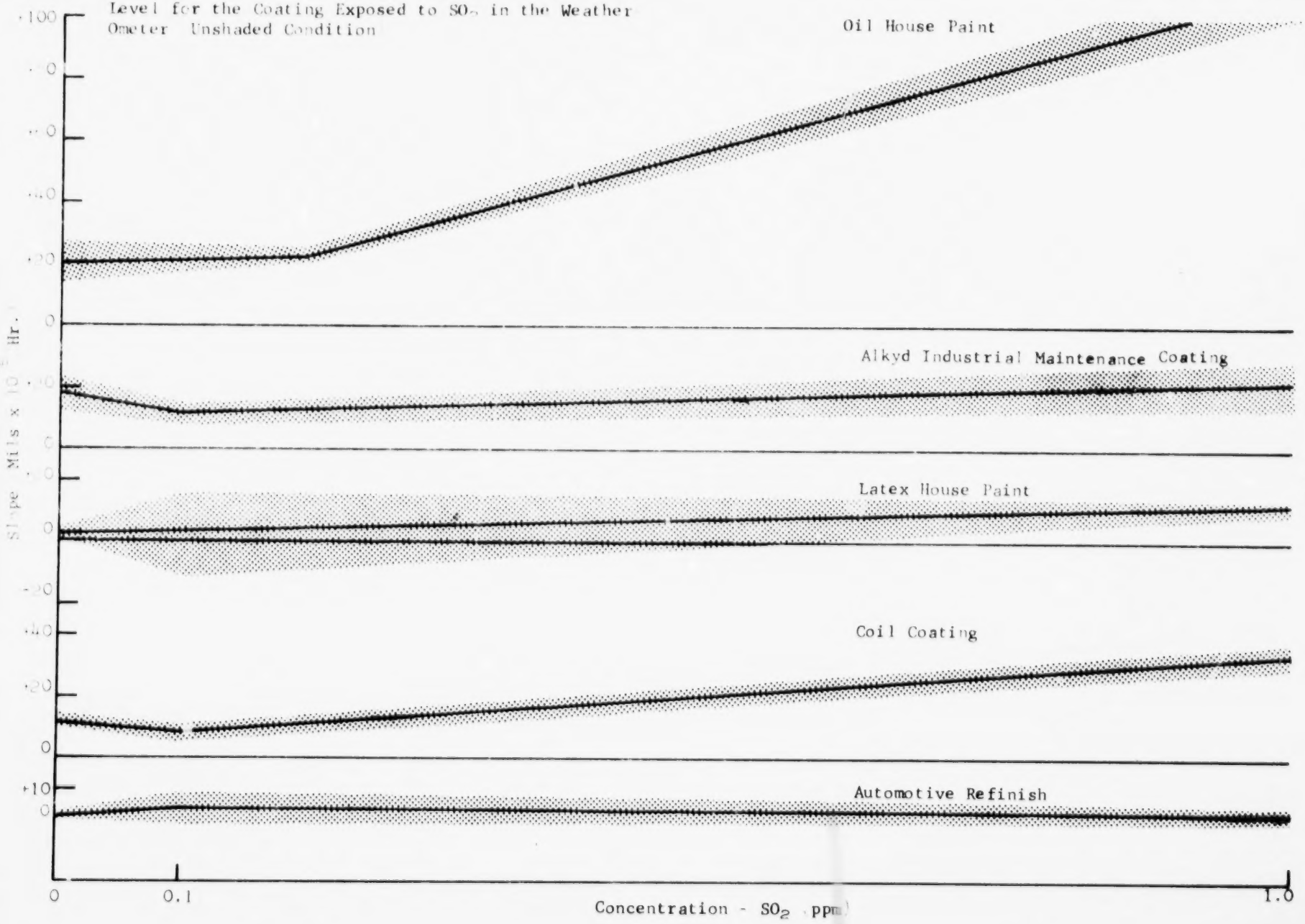


Figure . . - Slopes of Erosion Data Versus Pollutant Level for the Coatings Exposed to O_3 in the Weather Ometer Shaded Condition

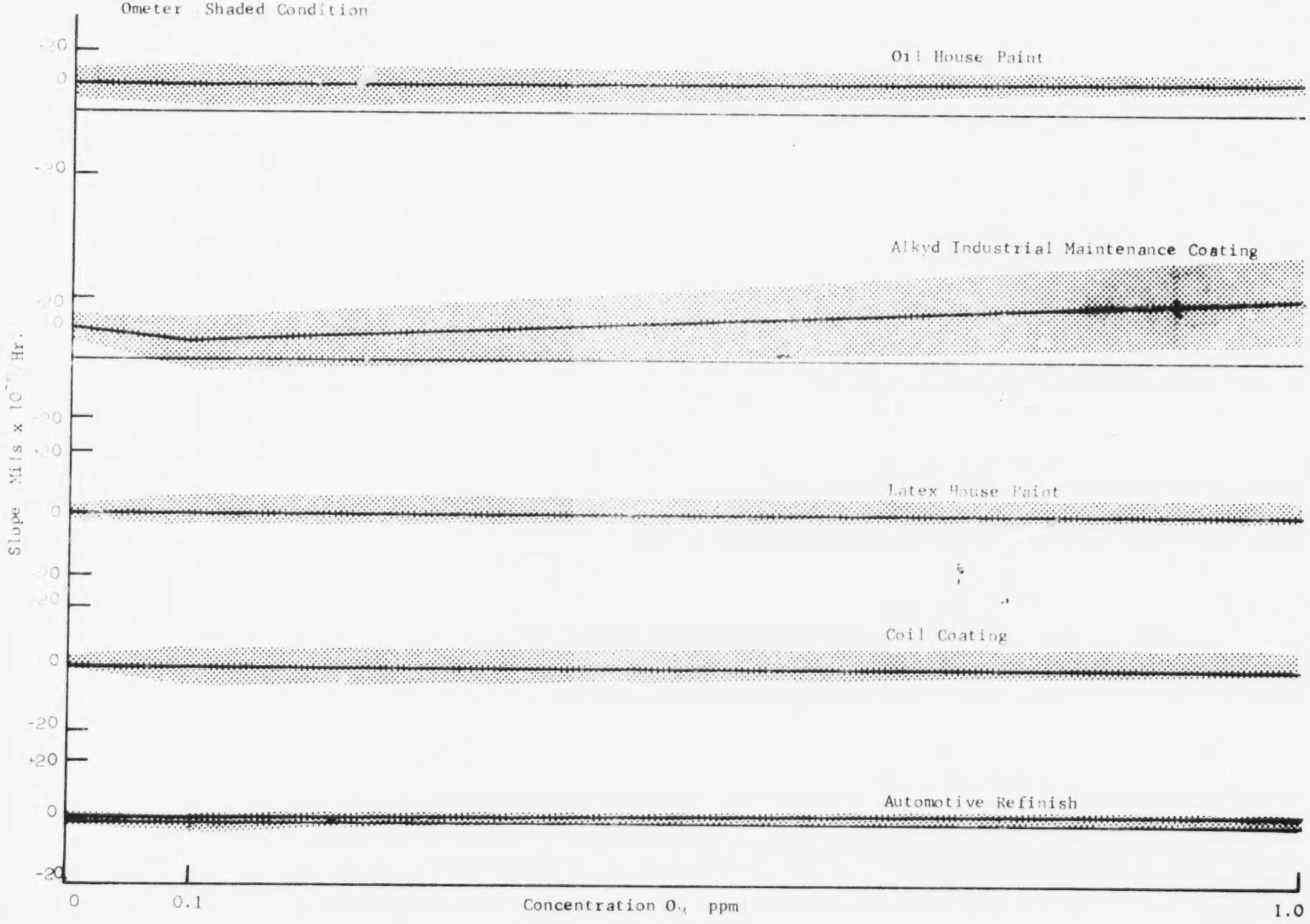


Figure 20 - Slopes of the Erosion Data Versus Pollutant Level for the Coatings Exposed to O_3 in the Weather-Ometer Unshaded Condition

It is interesting to note that despite the high statistical significance for the coatings affected by either SO_2 or O_3, the slope for the majority of coatings exposed to SO_2 at the 1.0 ppm level are generally greater than the slopes for the respective O_3 exposed coatings. This trend in results not only supports the contention that the greater erosion encountered at Chicago and Valparaiso versus Los Angeles and North Dakota is attributable to the SO_2 level but demonstrates conclusively that atmospheric pollutants under the given environmental conditon in the Weather-Ometer exert an adverse effect on the performance of specific coatings. It is also significant from the standpoint of correlation that the ranking in terms of erosion rates (slope) for the five coatings exposed in the Weather-Ometer was virtually the same as the ranking in terms of mils loss reported for the exterior studies. This consistency in trends in data indicate that exposure studies employing laboratory controlled environments can be used effectively in projecting pollutant damage of coatings exposed in exterior conditions.

(b) Infrared Analysis

ATR (Attenuated Total Reflectance) infrared measurements were conducted on the selected coatings after 1000 hours of exposure to the zero pollutant, 1 ppm SO_2 and 1 ppm O_3 levels. The results of this study showed that the oil house paint, the coil coating and the industrial maintenance coating degrade much more readily than the two acrylic systems (latex paint and automobile refinish) when exposed to 1 ppm SO_2 for 1000 hours in the Weather-Ometer. In the presence of ultraviolet light (unshaded condition), the oil/coating showed severe breakdowns of the binder, followed by the alkyd, urea/alkyd, acrylic latex and nitrocellulose/acrylic coatings, respectively. The shaded coatings exhibited a similar trend but with less binder degradation.

Examination of the 1.0 ppm O_3 samples after 1000 hours exposure showed spectra similar to the zero pollutant results but with slightly greater binder degradation. The extent of degradation was greater for the unshaded exposure (as expected) than for the shaded condition with the difference considerably less than that for the SO_2 exposed coatings (excluding the automotive refinish). In the case of the latter finish, the organic portion was considerably degraded by O_3 compared to the zero pollutant sample. In general, the above results are consistent with the trends reported for the exterior and Weather-Ometer erosion data (mils loss).

(c) Gloss and Sheen Measurements

All coatings were characterized by gloss and sheen during each examination period (400, 700 and 1000 hours) for the five pollutant conditions. However, only the gloss data is presented due to the inconsistencies exhibited by the sheen results. The cumulative gloss data presented in Table 15 show that only the automotive refinish and the industrial maintenance coating exhibit discernible trends between pollutant type and level. The graphs of gloss versus ppm SO_2 or O_3 levels for the latter two coatings presented in Figures 26 and 27 show that O_3 at the 0.1 ppm level for both shaded and unshaded conditions produced a considerable reduction in gloss particularly for the automotive refinish compared to SO_2. Consistently lower gloss was caused by exposure to

TABLE 15

Gloss Measurements (60°) of Panels Exposed to the Various Pollutant Conditions in the Weather-Ometer

SHADED EXPOSURE

		Zero Pollutant			0.1 ppm SO_2			1.0 ppm SO_2			0.1 ppm O_3			1.0 ppm O_3		
	Controls	400 Hr.	700 Hr.	1000 Hr.	400 Hr.	700 Hr.	1000 Hr.	400 Hr.	700 Hr.	1000 Hr.	400 Hr.	700 Hr.	1000 Hr.	400 Hr.	700 Hr.	1000 Hr.
Automotive Refinish	90	73	82	78	81	76	75	68	63	62	78	31	31	84	71	71
Industrial Maintenance	37	23	19	18	26	20	15	19	13	11	22	20	20	21	9	8
Oil House Paint	13	7	5	3	3	3	3	6	5	3	4	3	3	6	4	3
Latex House Paint	3	3	2	2	3	3	3	3	3	3	3	2	2	3	3	3
Coil Coating	5	3	3	2	4	3	2	3	3	3	3	3	2	2	2	2

UNSHADED EXPOSURE

Automotive Refinish	90	71	81	77	81	70	65	74	70	68	72	21	21	74	55	35
Industrial Maintenance	37	11	14	14	26	20	15	10	14	12	11	5	6	11	12	10
Oil House Paint	13	6	5	3	5	5	5	3.5	Failed		4	3	3	3	3	3
Latex House Paint	3	3	2	2	3	2	2	3	2	2	3	2	2	2	2	2
Coil Coating	3	2	2	3	3	3	3	2	2	3	3	2	2	2	2	2

Figure 26 Gloss Versus PPM SO_2 or O_3
After 1000 hours Exposure in the
Weather-Ometer Shaded Condition

Automotive Refinish ⊙
Industrial Maintenance ▽

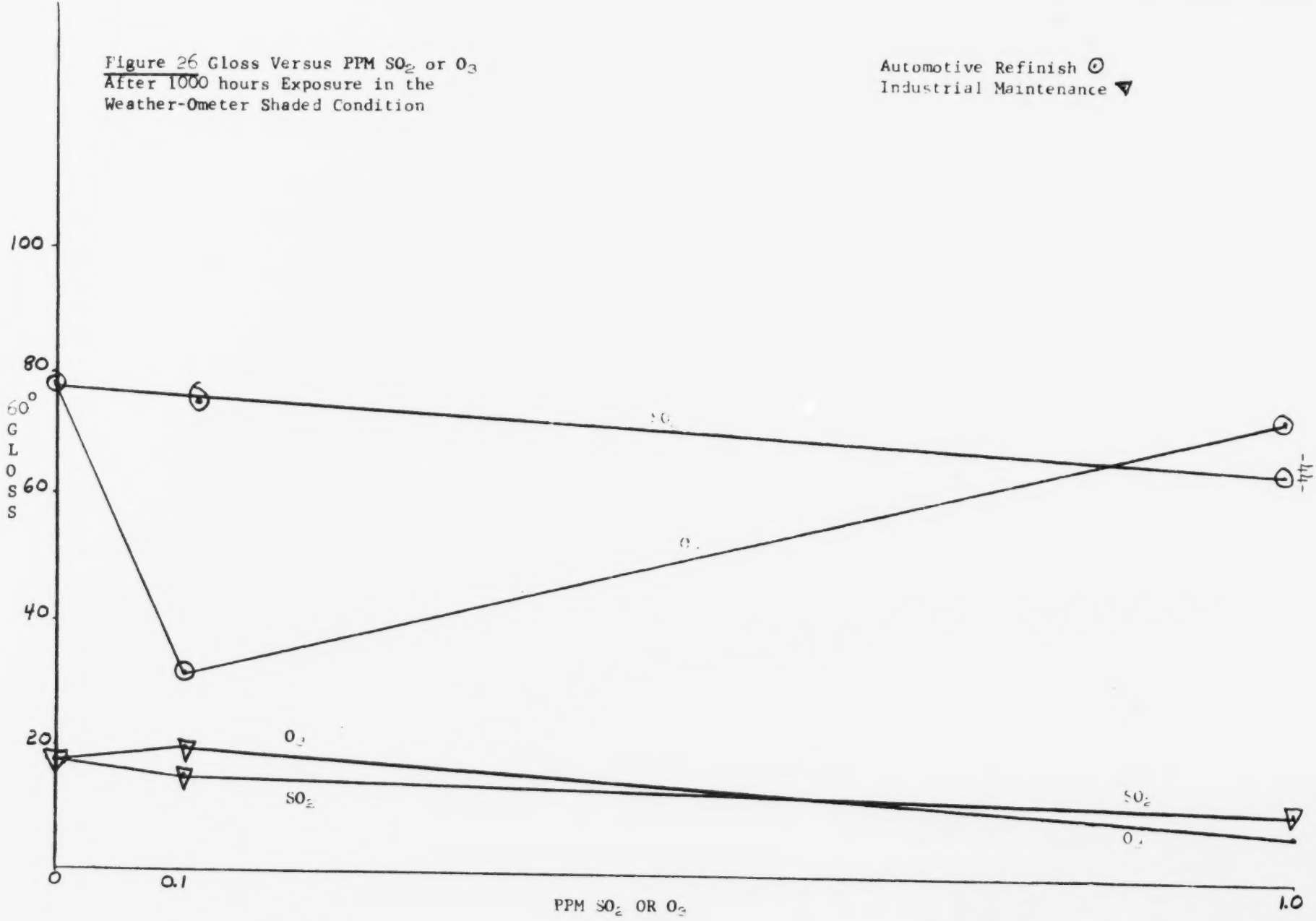
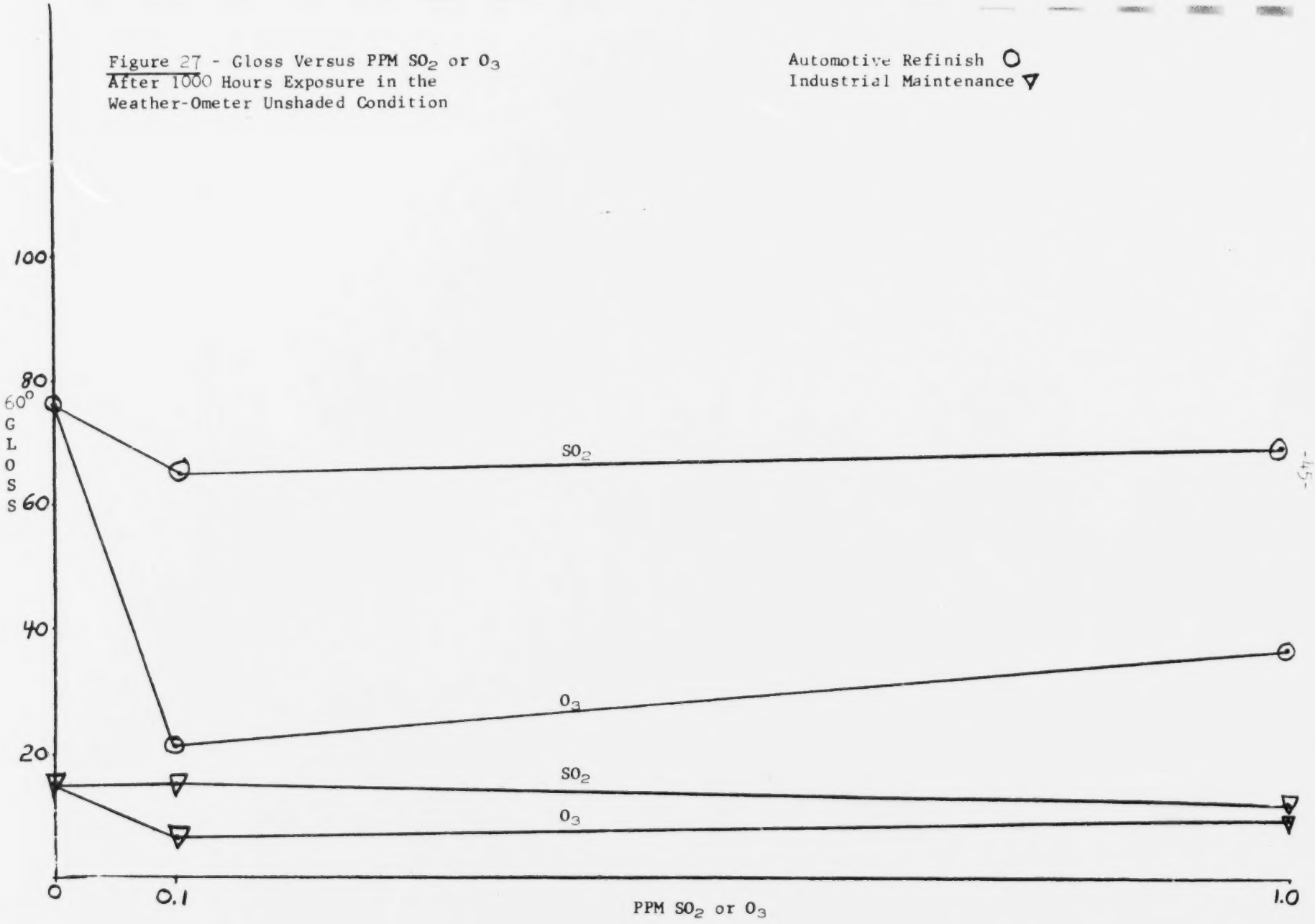


Figure 27 - Gloss Versus PPM SO_2 or O_3
After 1000 Hours Exposure in the
Weather-Ometer Unshaded Condition

Automotive Refinish ○
Industrial Maintenance ▽



O_3 at either ppm level in the unshaded condition. A similar but less pronounced trend is apparent for the alkyd industrial maintenance coating exposed in the unshaded condition.

As will be revealed later, the drastic drop in gloss of the automotive refinish is attributable to the considerable formation of water insoluble surface deposits of unknown composition on the panel surface. It is conceivable that the binder of the automotive refinish was degraded as indicated by the ATR spectra coincident to the formation of the surface deposits. These counterbalancing effects would reflect only a small but misleading weight loss. The latter contention would explain the apparent negligible effect of O_3 (0.1 or 1.0 ppm levels) on the erosion data (mils loss) for this coating. The above discussion illustrates the important point that more than one test method is required to accurately determine the effect of aggressive environments on coating performance. Also, the crystalline deposits noted on the O_3 exposed panels tends to obscure the validity of the gloss level being lower than the acceptable service life limit of 40.

(d) Surface Roughness Measurements

The surface roughness data collected on the coatings exposed to all five pollutant conditions in the Weather-Ometer (both north and south conditions) were assembled in tabular form (Table 16) and then ranked similarly in procedure as discussed under this section in Task II.

Table 17 presents the results of the rankings both within and between pollutant conditions (shaded and unshaded) for the respective coatings. Examination of the magnitude of the summation in columns shows that the oil house paint exhibited the greatest percentage change in surface roughness (low total value) with respect to the unexposed control, followed by the automotive refinish, coil coating, alkyd industrial maintenance and latex coating in descending order of severity. The above trends exist for both shaded or unshaded conditions irrespective of the pollutant type or level.

The effect of pollutant type and level is determined by summing the ranked value in rows. As before, a low summation is indicative of a large percentage change in surface roughness. The tabulated results for the shaded condition show that the zero pollutant exposure caused the least change in surface roughness with th 0.1 SO_2, 1.0 ppm SO_2, 0.1 ppm O_3 and 1.0 ppm O_3 producing increasing percentage change in roughness, respectively. A similar analysis for the unshaded coatings is precluded because of the failure of the coatings exposed to 1.0 ppm SO_2.

The trends in rankings are again inconsistent with the erosion results collected in Task II and III in that high surface roughness is not equated with high erosion. However, ozone at either the 0.1 or 1.0 ppm level produced the greatest effect on surface roughness as well as the previously discussed detrimental effect on gloss.

TABLE 16

Surface Roughness (Percent Change with Respect to Control) of Weather-Ometer Panels After 1,000 Hours Exposure at Various Pollutant Levels

	Shaded					Unshaded				
	"0" Pollutant	0.1 ppm SO_2	1 ppm SO_2	.1 ppm O_3	1 ppm O_3	"0" Pollutant	0.1 ppm SO_2	1 ppm SO_2	.1 ppm O_3	1 ppm O_3
Oil House Paint	58	39	55	60	71	53	34	-	42	58
Automotive Refinish	23	15	31	23	150	23	15	31	23	92
Coil Coating	0	24	17	29	17	-12	17	-4.8	17	-14
Industrial Maintenance	-9.3	-4.6	-2.3	6.9	4.6	30	-25	-12	6.9	-6.9
Latex House Paint	-15	3.4	-4.5	3.4	6.7	-25	1.1	1.1	0	3.4

TABLE 17

Ranking of Percent Change in Surface Roughness of Films Exposed in Weather-Ometer at Various Pollutant Levels

COATINGS

Pollutant Level	Ranking Within Pollutants - Shaded				
	Automotive Refinish	Latex Coating	Coil Coating	Industrial Maintenance	Oil House Paint
"0" Pollutant					1
0.1 ppm SO_2					1
1.0 ppm SO_2					1
0.1 ppm O_3					1
1.0 ppm O_3	1				2
Summation	11				

	Ranking Within Pollutants - Unshaded				
	Automotive Refinish	Latex Coating	Coil Coating	Industrial Maintenance	Oil House Paint
"0" Pollutant					1
0.1 ppm SO_2					1
1.0 ppm SO_2					1
0.1 ppm O_3					1
1.0 ppm O_3	1				2
Summation	11				

	Ranking Between Pollutants - Shaded					Sum.
	Automotive Refinish	Latex Coating	Coil Coating	Industrial Maintanance	Oil House Paint	
"0" Pollutant						21.5
0.1 ppm SO_2						18.5
1.0 ppm SO_2						16.5
0.1 ppm O_3			1	1		10.0
1.0 ppm O_3	1	1			1	8.5

	Ranking Between Pollutants - Unshaded					
	Automotive Refinish	Latex Coating	Coil Coating	Industrial Maintenance	Oil House Paint	Sum.
"0" Pollutant						15.5
0.1 ppm SO_2						15.0
1.0 ppm SO_2					Failed	--
0.1 ppm O_3						14
1.0 ppm O_3	1	1			1	11

(e) Instron Determinations

Stress-strain analyses performed on free films of the selected coatings after zero, 400 and 1000 hours of exposure to the various pollutant conditions generally yielded inconclusive results. As shown in Table 18 virtually all coatings (excluding the latex paint) exhibited inconsistent trends in peaks in tensile strength within and between the shaded and unshaded conditions. Some of these abnormalities may have been resolved through additional data points. However, this latter limitation may not be of practical significance since the automotive refinish, coil coating and oil house paint became relatively brittle (approximately 2% or less elongation) after 400 hours of exposure (shaded and unshaded conditions). As noted previously, tensile strength measurements of brittle coatings are difficult to reproduce with good precision. Consequently, any apparent trends in results for these coatings would be fortuitous. In the case of the flexible latex coating, it is noted that an inflection point in tensile strength occurred only at 1.0 ppm SO_2 (shaded condition). All other exposures caused an expected increase in tensile strength with the level being higher in the unshaded condition.

(f) Sorption-Desorption Measurements

The coatings exposed for 1000 hours to the zero, 1.0 ppm SO_2 and 1.0 ppm O_3 pollutant levels in the Weather-Ometer (both shaded and unshaded) along with the appropriate unexposed controls were characterized in terms of their sorption-desorption properties. One objective of this auxillary project was to determine whether sorption-desorption measurements of the unexposed control coatings could be used to predict the susceptibility to attack particularly by SO_2 during subsequent exposure in the Weather-Ometer studies. It was reasoned that coatings which retain a moderate to high level of moisture following exposure to a specified desorption condition would be more readily affected by exposure to SO_2 containing environments. The greater coating degradation (erosion) expected would result from longer exposure to the hydrolytic action of dilute sulfurous acid formed from absorbed SO_2 and the water within the film. In addition to the above objective, it was desired to determine the magnitude of change in sorption-desorption properties with respect to those of the unexposed control that might occur during exposure to the various pollutant conditions (zero, 1 ppm SO_2 or O_3) in the Weather-Ometer.

Figures 27 through 32 show graphs of the normalized weight gain (sorption) or weight loss (desorption) in milligrams versus the respective periodic weighing. Essentially all of the coatings irrespective of the pollutant type or level achieved equilibrium sorption or desorption (constant weight) within a 2-6 hour period. The rapid equilibration of the unexposed control paints (Figure 28) during the desorption period nullifies the hypothesis of predicting the susceptibility of attack by SO_2 through initial sorption-desorption measurements.

Examination of the data for the exposed coatings in Figures 29-32 shows that pollutants irrespective of the type or level effected an insignificant change (2-4 milligrams) in the equilibrium sorption level for any given coating. Only the latex paint exhibited a marked decrease in sorption level compared to the unexposed control results (Figure 28). The relationship of sorption and erosion

Figure [illegible] - Sorption - Desorption of Unexposed Controls

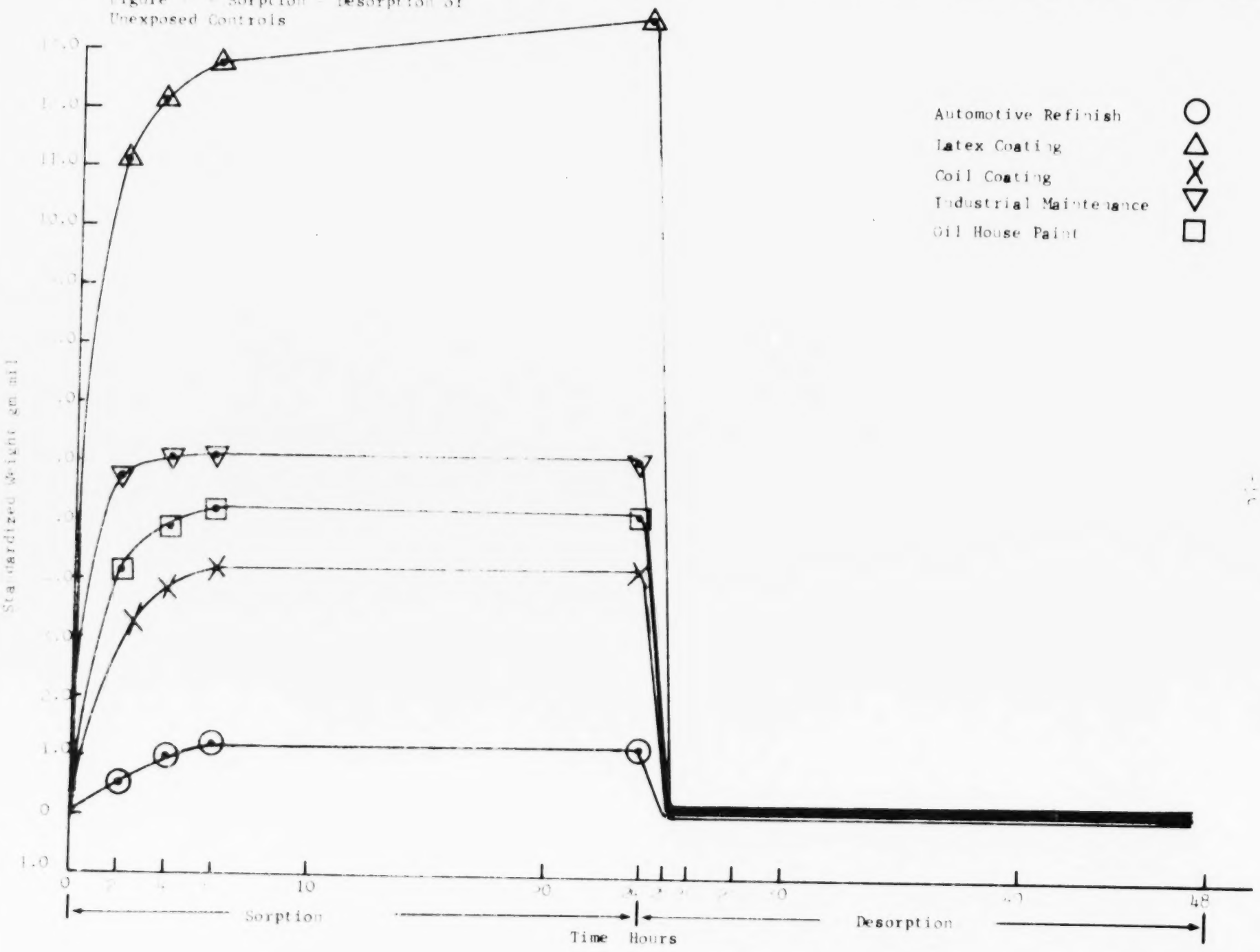


Figure 29 - Sorption-Desorption of Weather-Ometer Panels Exposed to 1,000 Hours "0" Pollutants. Shaded

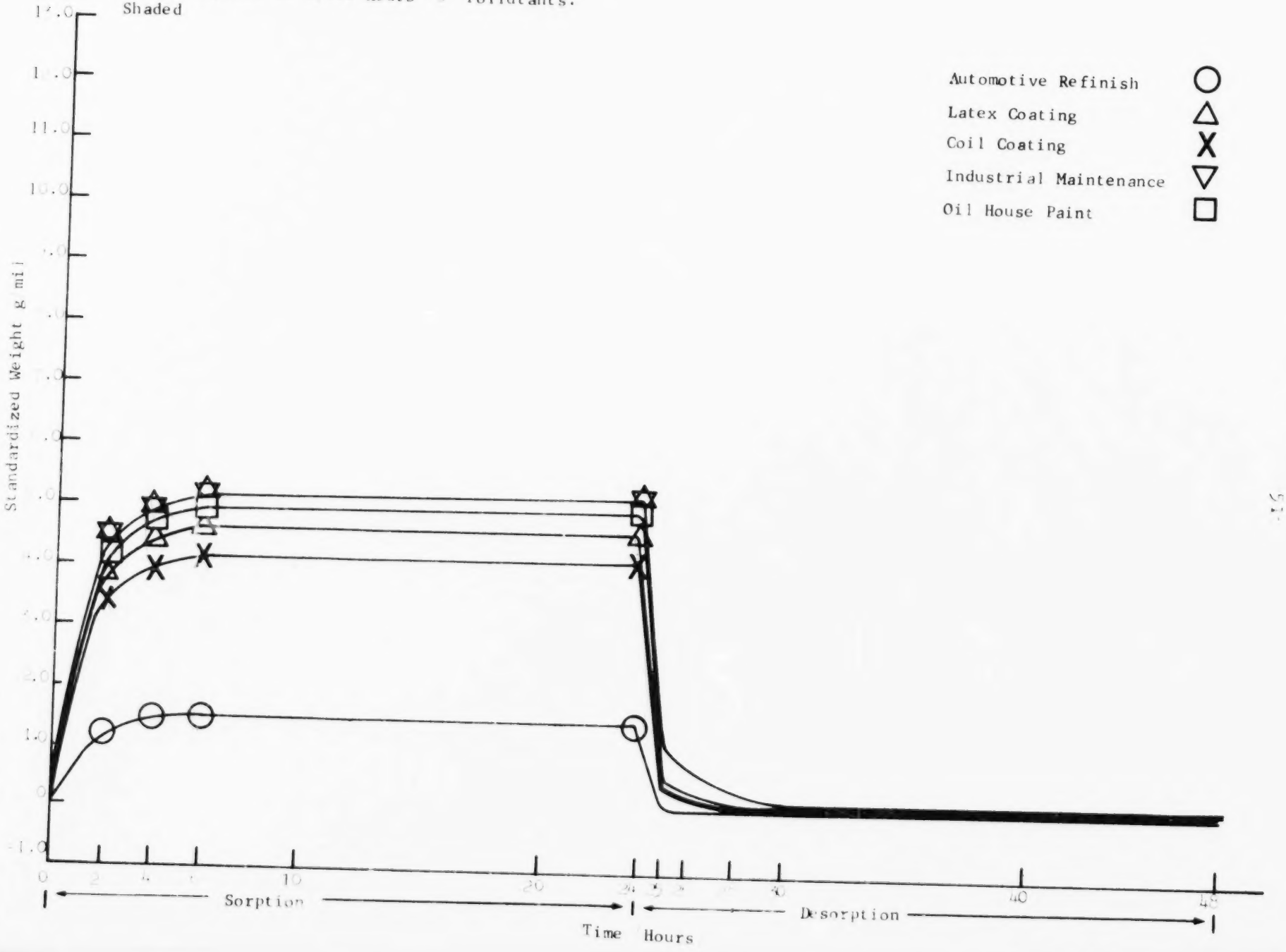


Figure 30 - Sorption-Desorption of Weather Ometer Panels Exposed for a 1,000 Hours to "O" Pollutants. Unshaded

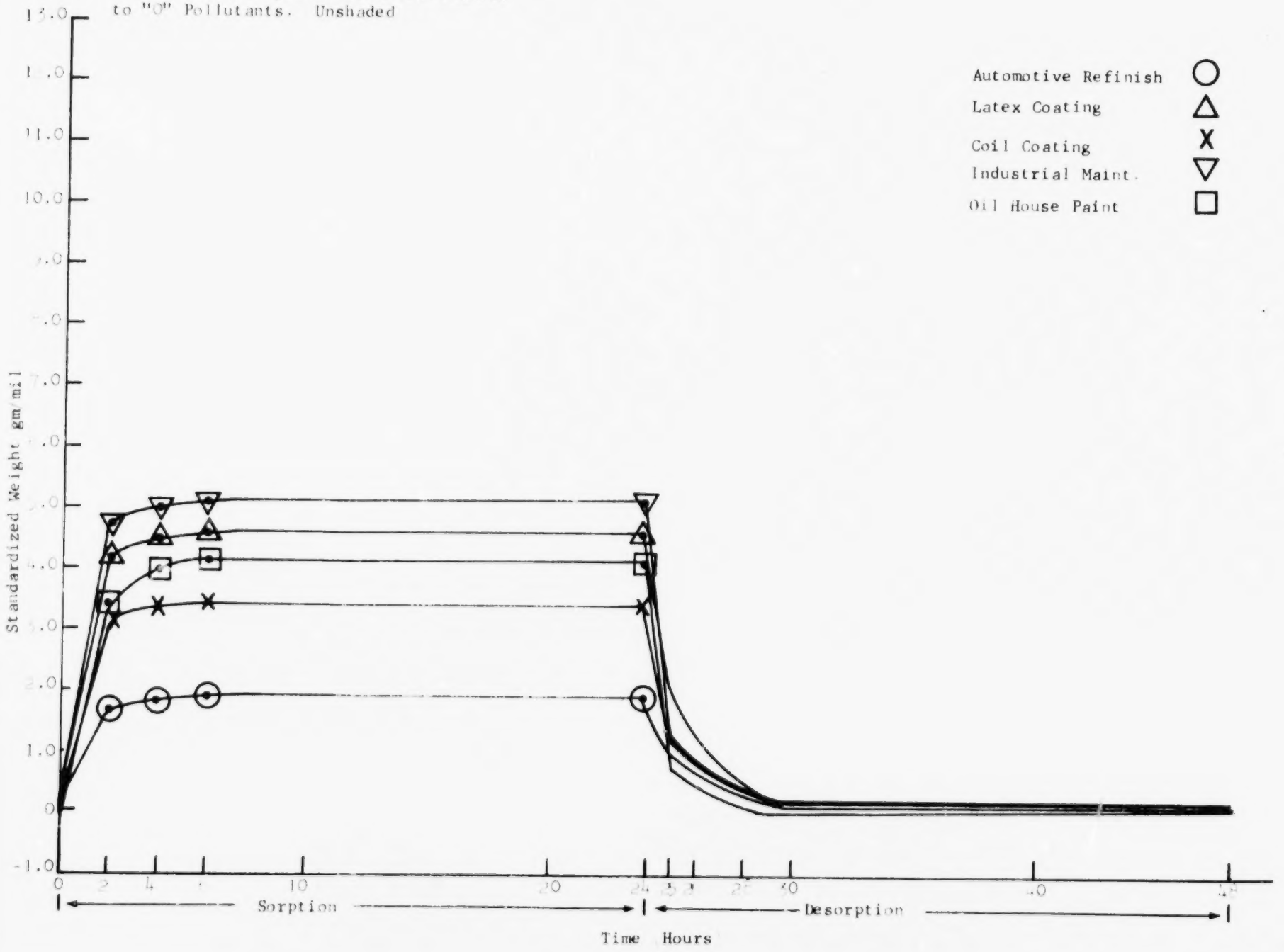


Figure 31 - Sorption-Desorption of Weather-Ometer Panels Exposed for 1000 Hours to 1.0 ppm O_3. Shaded.

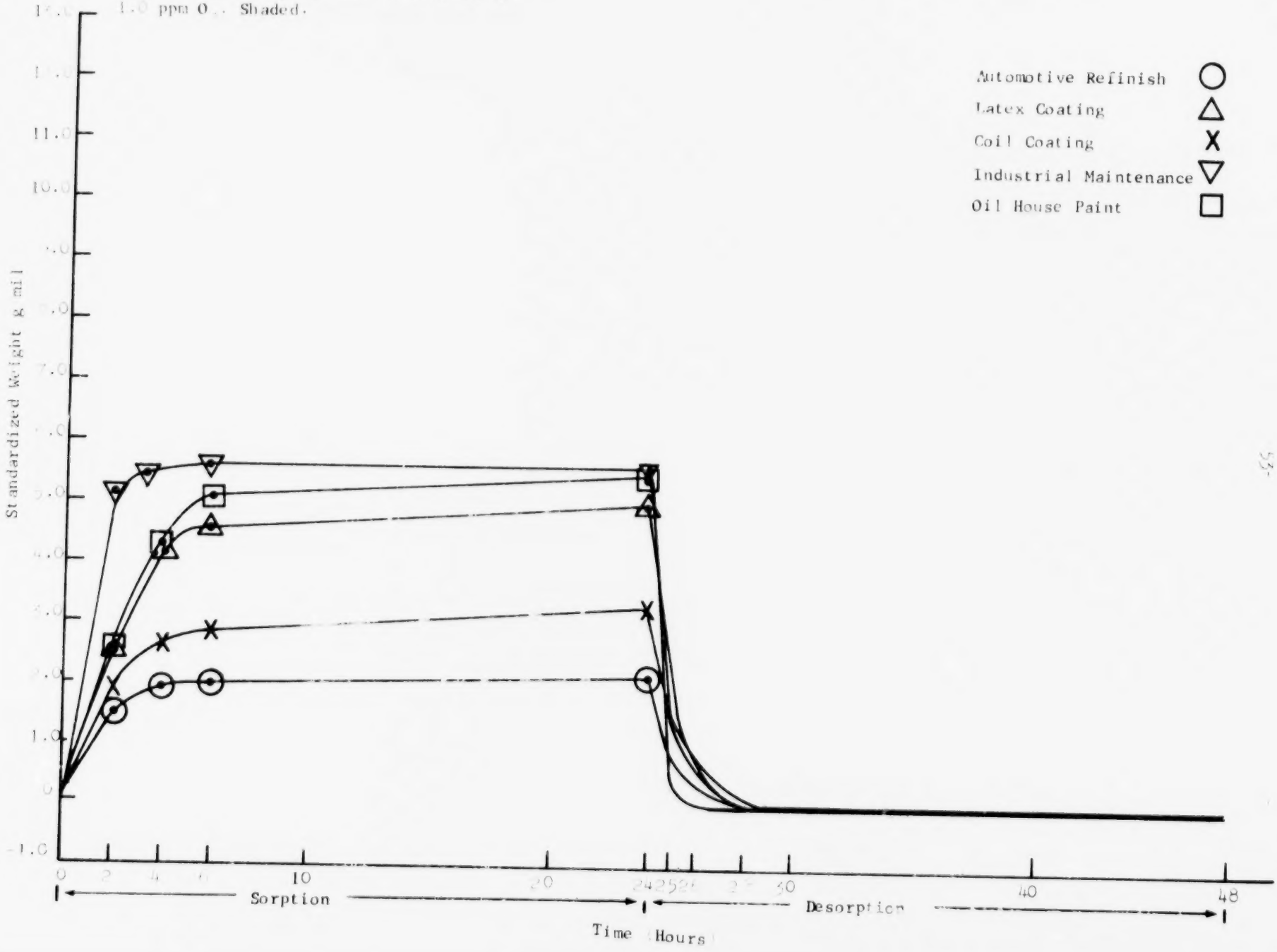


Figure 52 - Sorption-Desorption of Weather-Ometer Panels Exposed for 1000 hours to 1 ppm O_3. Unshaded.

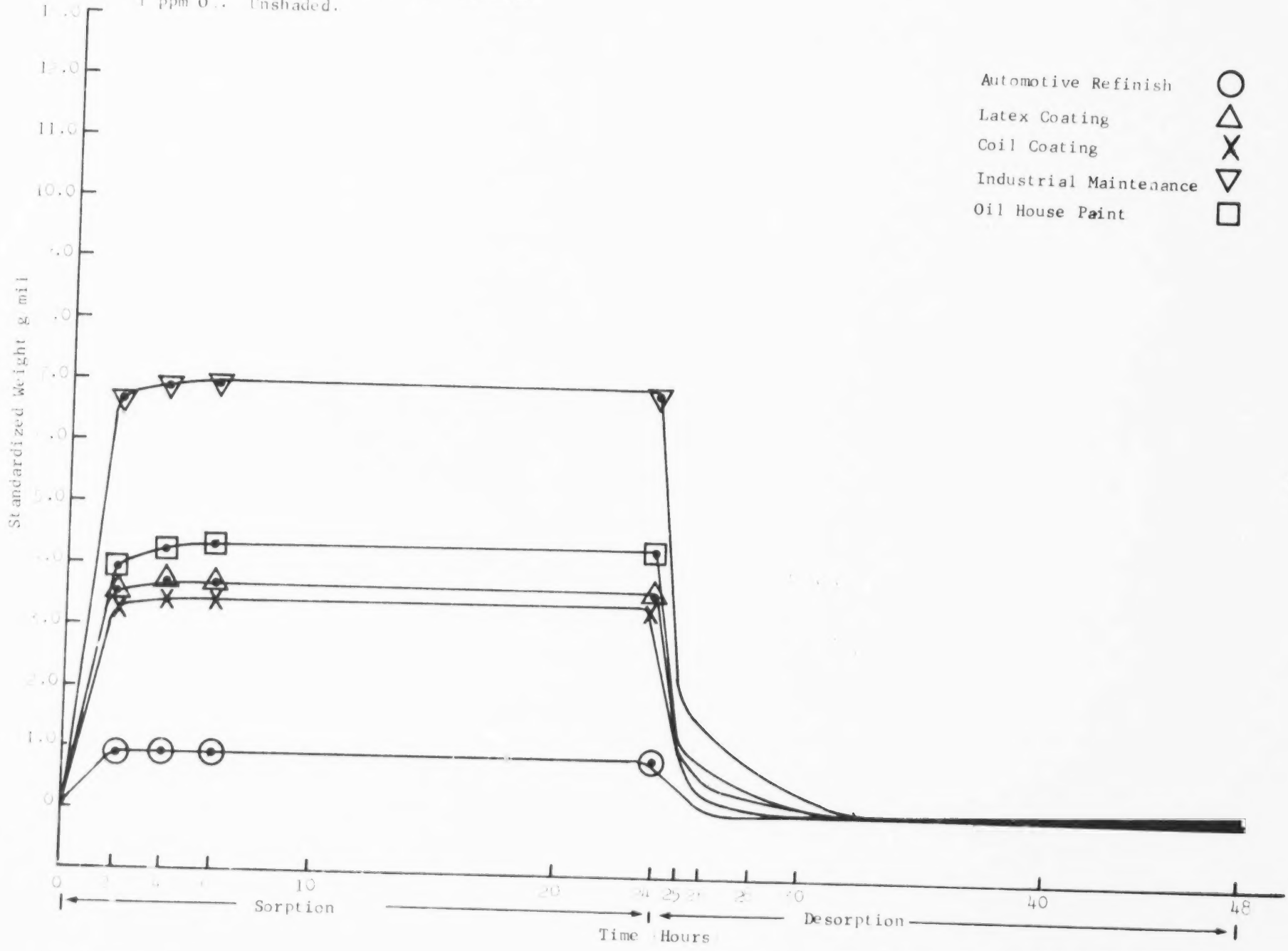


Figure 33 - Sorption-Desorption of Weather-Ometer Panels Exposed for a 1,000 Hours to 1.0 ppm SO_2 Shaded

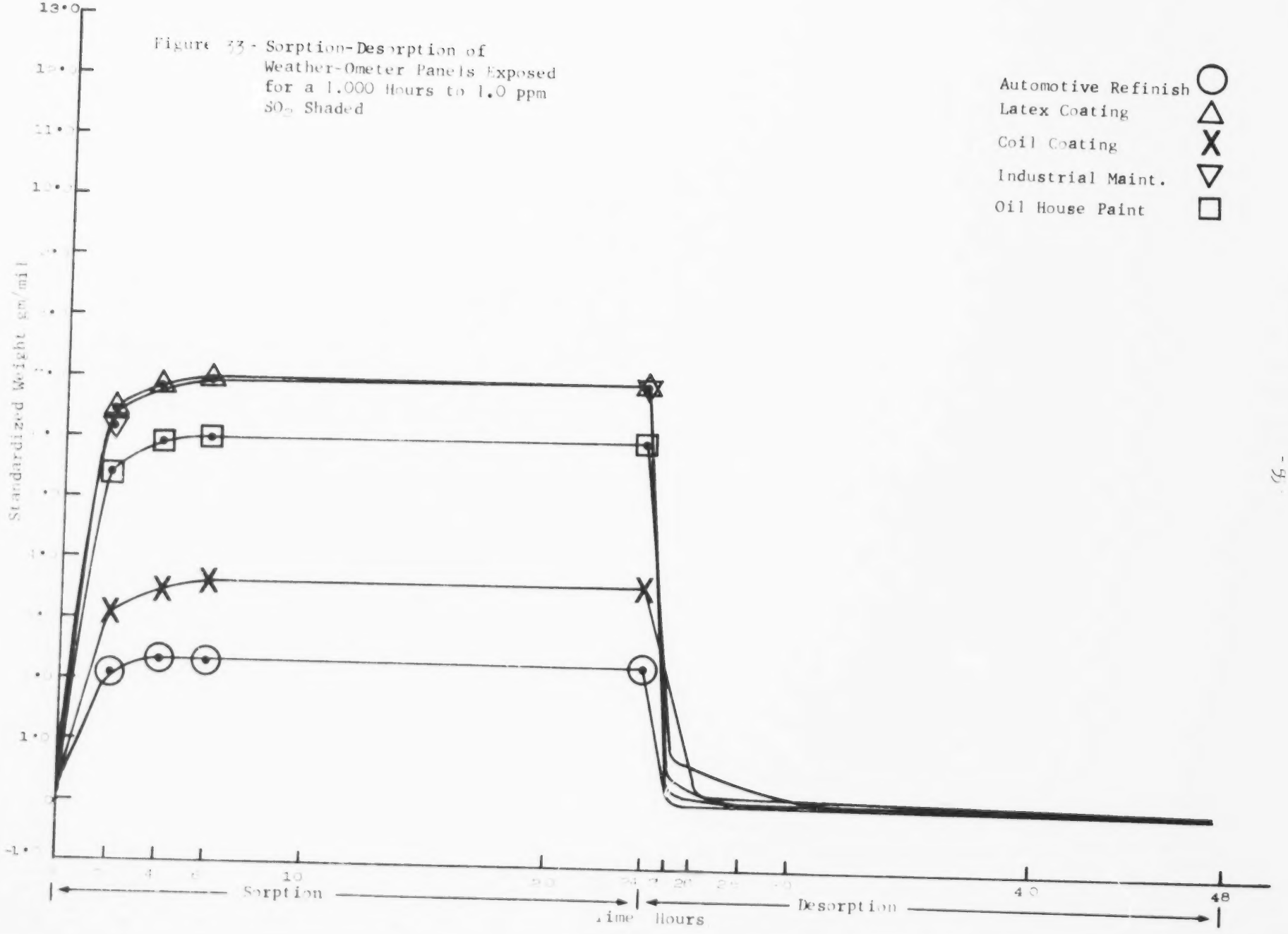


Figure 4 - Sorption-Desorption of Weather-Ometer Panels Exposed for 1,000 Hours to 1.0 ppm SO_2 Unshaded

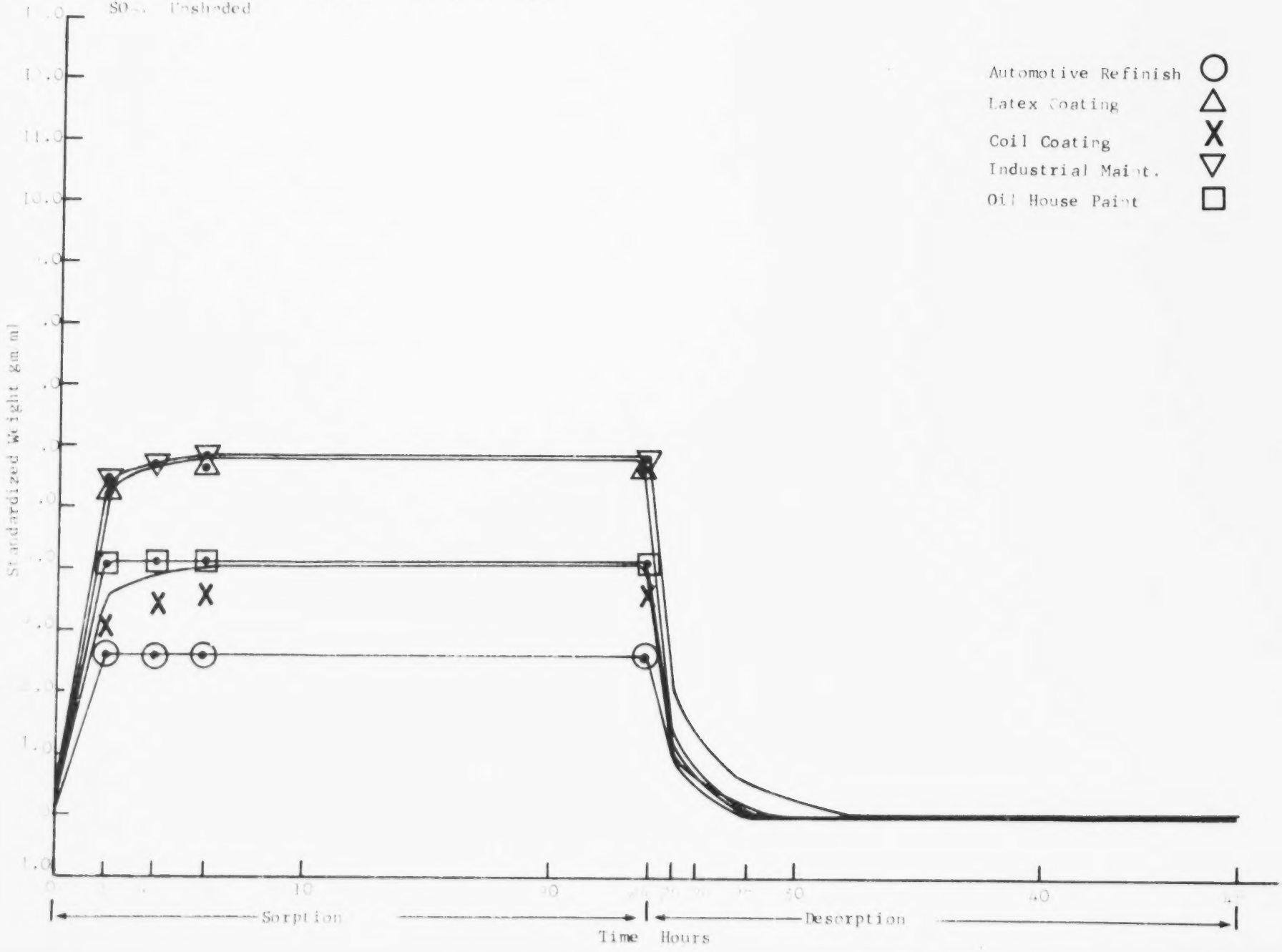


TABLE 18

Tensile Strength Determinations (PSI) of Films Exposed in Weather-Ometer at Various Pollutant Levels

SHADED

	Controls	"0" Pollutant		0.1 ppm SO_2		1 ppm SO_2		0.1 ppm O_3		1 ppm O_3	
		400 (Hr.)	1,000 (Hr.)	400 (Hr.)	1,000 (Hr.)	400 (Hr.)	1,000 (Hr.)	400 (Hr.)	1000 (Hr.)	400 (Hr.)	1000 (Hr.)
Automotive Refinish	2800	3050	2850	3150	3400	2800	2550	3450	2950	3200	1350
Latex Coating	850	1500	1450	1500	1700	1550	1400	1700	2050	1300	1900
Coil Coating	1670	1650	2000	1800	2550	2300	2050	1700	1700	1750	2050
Industrial Maintenance	500	1400	1650	1850	1350	1600	1250	1700	3000	1150	1700
Oil House Paint	400	1150	900	1250	1050	900	1000	1150	1450	1300	1350

UNSHADED

Automotive Refinish	2800	3500	3200	2300	1850	1750	1600	3150	2850	3200	1250
Latex Coating	850	2000	2300	1750	2100	1300	2100	1800	2500	2050	2250
Coil Coating	1670	2600	1900	1350	3150	1250	1600	2200	2150	1650	2200
Industrial Maintenance	500	1950	1750	1550	2050	1400	2200	1600	2400	1550	1100
Oil House Paint	400	1150	1050	950	1400	800	1300	1550	1150	1050	900

Tensile Strength rounded to nearest 50 PSI.

is consistent only for the automotive refinish which shows the least sensitivity to moisture and the lowest level of erosion

The above discussion reveals that the technique of sorption-desorption is not a totally reliable indicator of the effect of aggressive environments on coating performance

g Scanning Electron Photomicrographs

Films of the five coatings exposed to the zero pollutant, 1.0 ppm SO_2 or 1.0 ppm O_3 conditions (unshaded only) in the Weather-Ometer were examined with the scanning electron microscope. Unexposed controls were also included in the series for comparison.

The photomicrographs presented in Figures through are arranged according to a coating type to facilitate discerning the extent of surface degradation caused by the three pollutant conditions. Two levels of magnification (700x, 7000x) were employed to enable a critical analysis of the topographical changes which occurred at the coating surface.

Examination of the unexposed controls in Figures , , , and show a layer of binder which obscures the underlying pigment morphology. Only the latex house paint and the coil coating exhibit a significant degree of initial surface roughness.

The level of degradation caused by exposure to the three pollutant conditions is discerned through examining the surface texture at 700x magnification and the pigment (TiO_2 and or extender) binder matrix at the 7000x level. Holes appearing in the coating surface indicate the loss of agglomerated pigment consisting of combinations of TiO_2, inert and/or binder. Large depressions or crevices between the relatively massive extender particles also suggest a greater degree of surface degradation.

In view of the above degradative features, it is apparent that the latex house paint, the coil coating and the oil house paint (Figures to , to and to respectively) show the greatest amount of physical change in respect to the unexposed control during exposures to the three pollutant conditions. Exposures to 1 ppm SO_2 appears to have effected the greatest degradation which diminishes on exposure to 1 ppm O_3 and the zero pollutant levels, respectively. Dramatic differences in surface topography are evident between the 1 ppm SO_2 and the zero pollutant levels for the latex coating and the oil house paint. The surface of the latter two coatings (7000x) exposed to the zero pollutant condition show a considerable amount of apparently unaffected binder.

Although the film of the oil house paint (Figure 7000x) exposed to the 1.0 ppm SO_2 condition exhibited major surface modification as evidenced by the deep fissures between the massive extender particles, even greater degradation on a macroscopic level occurred during similar exposure of the coated metal panels. Extensive microchecks developed randomly over the latter coating surface between 400 and 600 hours of exposure. The reflected light micrograph

presented in Figure [illegible] shows a portion of the failure which extends almost to the metal substrate.

Examination of the alkyd industrial maintenance coating at 700x magnification (Figures [illegible] and [illegible]) shows a random development of holes in the surface of all exposed samples. However, the surface topography revealed in the 7000x photographs is relatively uniform with no apparent difference in pigment packing irrespective of the three pollutant conditions.

The relatively non-descript surface of the automotive refinish (Figures 66 and 75) indicate that this coating was the least affected in respect to binder degradation during exposure to the three pollutant conditions. Exposure to 1.0 ppm SO_2 has apparently caused slightly greater surface roughness than exposure to the zero pollutant level. However, this observed topography change attributable to binder-pigment loss exerted an inconsequential effect on the erosion rate (Table I). The crystal formation evident on the surface of both the 0.1 ppm and 1.0 ppm O_3 exposed samples presented in Figure 70 and 71 (700x) explains the respective drastic reduction in gloss level noted in Table I and the increased surface roughness apparent in Table I. These deposits of unknown composition are more pronounced on the 0.1 ppm O_3 sample (Figure 70).

The above discussion clearly shows that SEM analyses provide a useful qualitative technique in characterizing the degradation of coatings exposed to aggressive environments. The photomicrographs show trends consistent with the erosion rate data (Table I) in respect to the greater degradation caused by SO_2 versus O_3 or the zero pollutant conditions. Comparison can readily be made between coatings showing extremes in degradation. However, difficulties are encountered in ranking the durabilities of coatings such as the latex house paint, coil coating and the oil house paint which contain extenders as part of the pigment composition and exhibit major apparent surface roughness due to exposure.

The scanning electron photomicrographs are consistent with the surface roughnss data (Table I) only for the automotive refinish, the oil house paint and possibly the latex paint. The coil coating shows decreased surface roughness irrespective of the three pollutant conditions but considerable surface degradation holes indicating loss of agglomerates according to the photomicrographs (Figures [illegible], [illegible], [illegible]). The general increase in apparent roughness of the exposed industrial maintenance coating versus the unexposed control (Figures [illegible], [illegible], [illegible], and [illegible] respectively) is also contrary to the surface roughness measurements.

LATEX HOUSE PAINT

Figure 35 - Unexposed Control . 700X

Figure 36 - " " 7000X

Figure 37 - 1000 Hours Exposure to Zero Pollutant - Unshaded 700X

Figure 38 - " " " " " " " 7000X

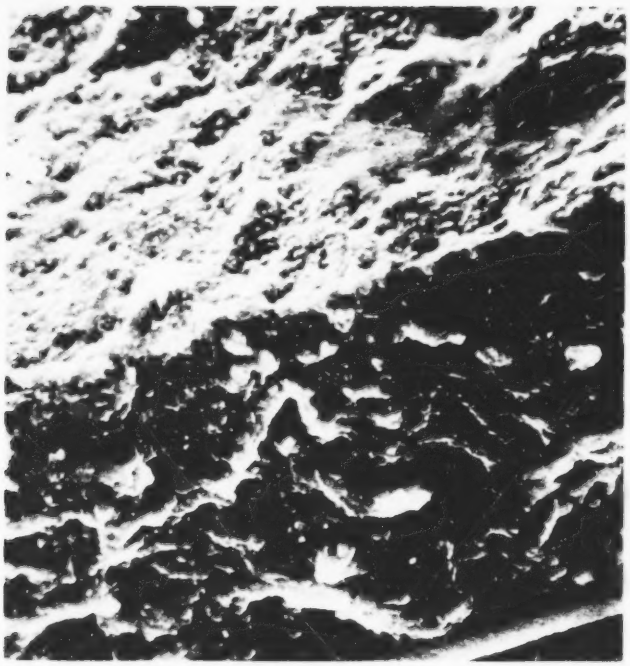


Fig. 35

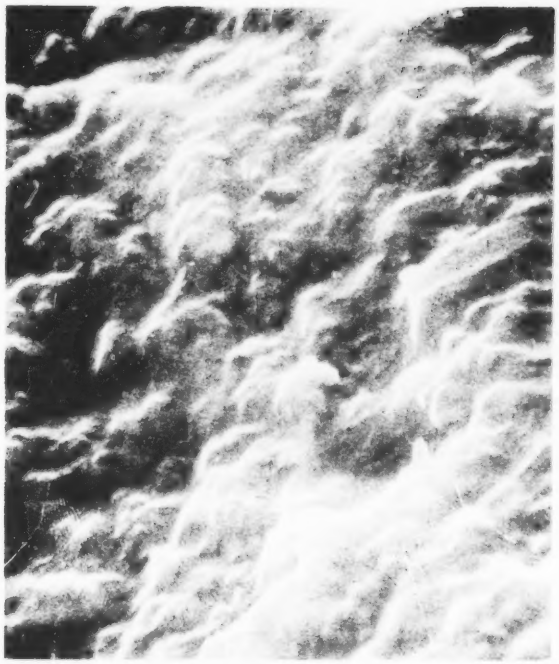


Fig. 36

Fig. 37

Fig. 38

LATEX HOUSE PAINT

Figure 39 - 1000 Hours Exposure to 1.0 ppm O_3 - Unshaded 700X

Figure 40 - " " " " " " O_3 - " 7000X

Figure 41 - " " " " " " SO_2 - " 700X

Figure 42 - " " " " " " SO_2 - " 7000X

Fig. 39

Fig. 40

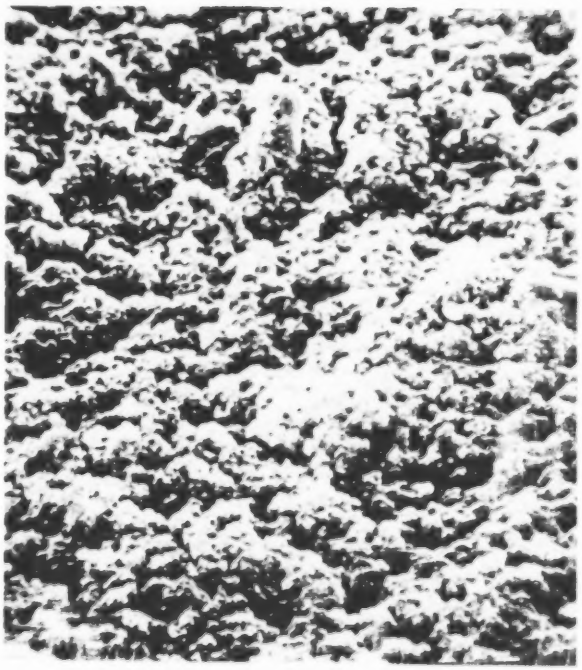


Fig. 41

Fig. 42

COIL COATING

Figure 43 - Unexposed Control 700X

Figure 44 - " " 7000X

Figure 45 - 1000 Hours Exposure to Zero Pollutant - Unshaded 700X

Figure 46 - 1000 " " " " " " 7000X

Fig. 43

Fig. 44

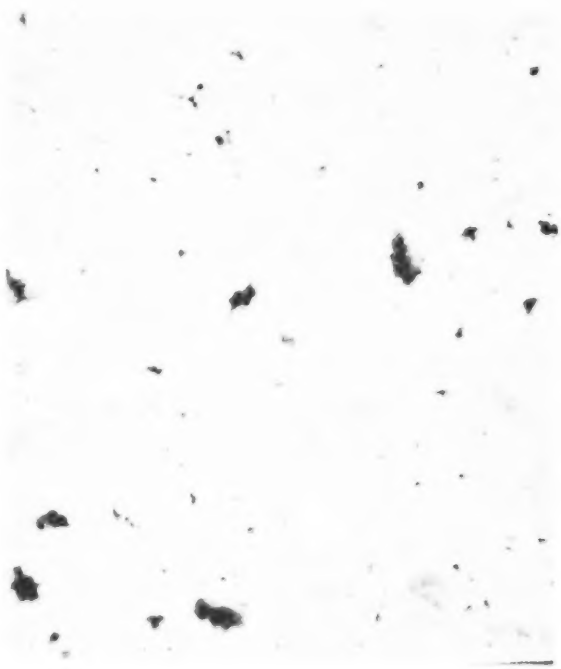


Fig. 45

Fig. 46

COIL COATING

Figure 47 - 1000 Hours Exposure to 1.0 ppm O_3 - Unshaded 700X

Figure 48 - " " " " " " " - " 7000X

Figure 49 - " " " " " " SO_2 - " 700X

Figure 50 - " " " " " " " - " 7000X

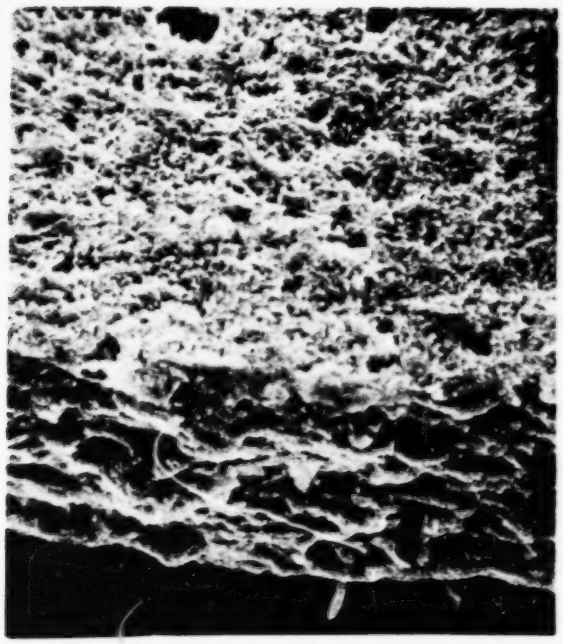


Fig. 47

Fig. 48

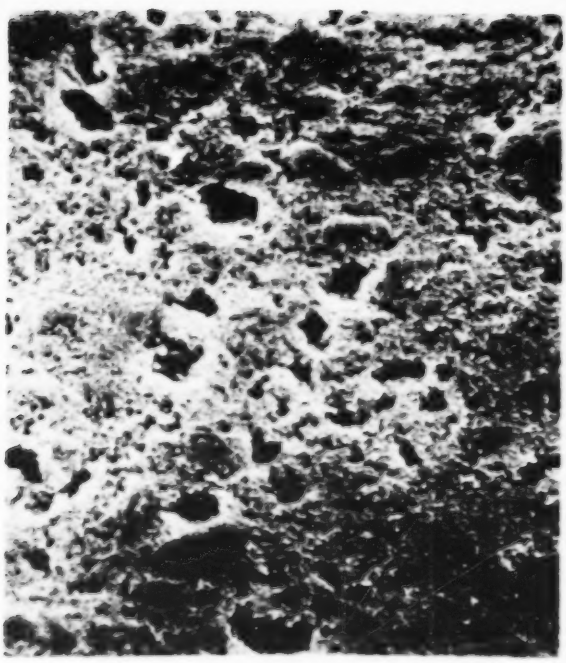


Fig. 49

Fig. 50

OIL HOUSE PAINT

Figure 51 - Unexposed Control 700X

Figure 52 - 1000 Hours Exposure to Zero Pollutant - Unshaded 700X

Figure 53 - " " " " " " " 7000X

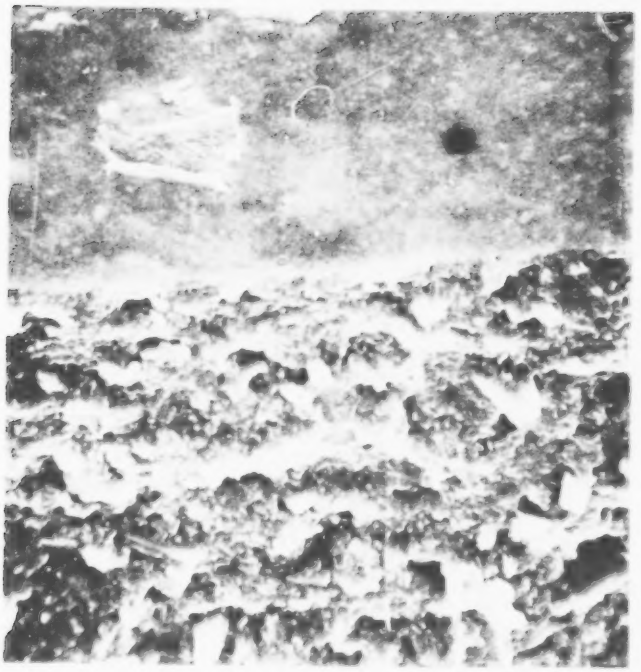


Fig. 51

Fig. 52

Fig. 53

OIL HOUSE PAINT

Figure 54 - 1000 Hours Exposure to 1.0 ppm O_3 Unshaded 700X

Figure 55 - 1000 " " " " " " " 7000X

Figure 56 - 1000 " " " " " SO_2 " 700X

Figure 57 - 1000 " " " " " " " 7000X

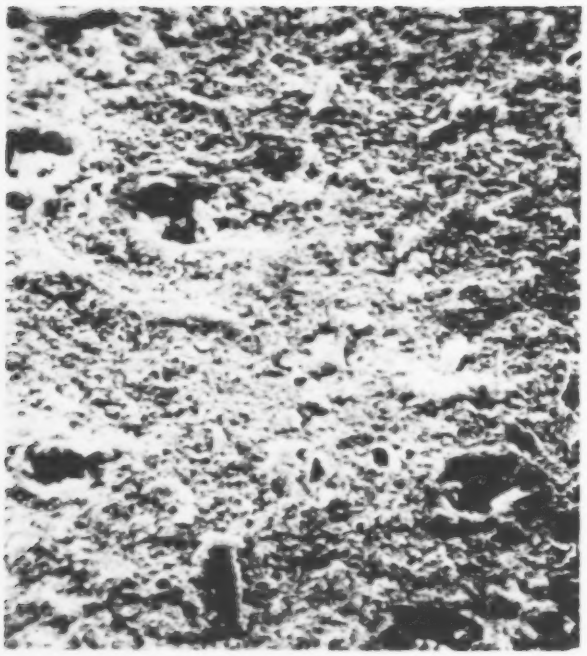


Fig. 54

Fig. 55

Fig. 56

Fig. 57

OIL HOUSE PAINT

Figure 58 - 1000 Hours Exposure to 1.0 ppm SO_2 Shaded 130X

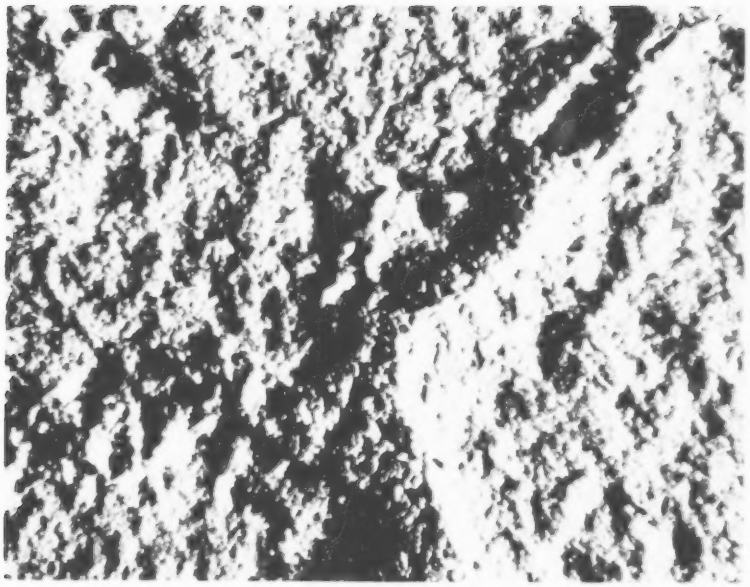


Fig. 58

INDUSTRIAL MAINTENANCE COATING

Figure 59 - Unexposed Control 700X

Figure 60 - 1000 Hours Exposure to Zero Pollutant 700X

Figure 61 - " " " " " " 7000X

Fig. 59

Fig. 60

Fig. 61

INDUSTRIAL MAINTENANCE COATING

Figure 2 - 1000 Hours Exposure to 1.0 ppm O_3 Unshaded 700X

Figure 3 - " " " " " " " " 7000X

Figure 4 - " " " " " " SO_2 " 700X

Figure 5 - " " " " " " " " 7000X

Fig. 62

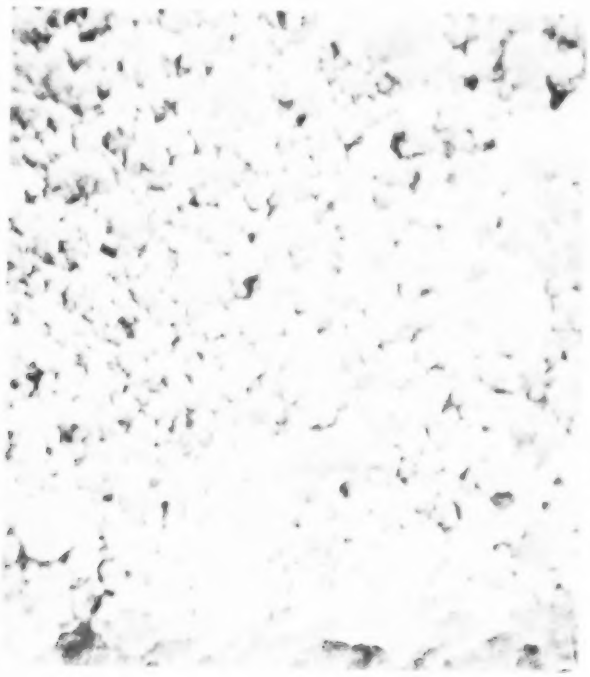


Fig. 63

Fig. 64

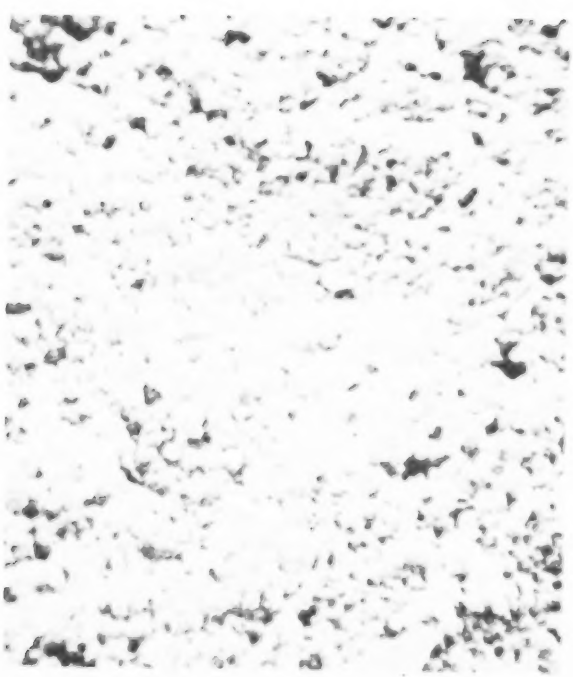


Fig. 65

AUTOMOTIVE REFINISH

Figure [illegible] - Unexposed Control 700X

Figure [illegible] - " " 7000X

Figure [illegible] - 1000 Hours Exposure to Zero Pollutant - Unshaded 700X

Figure [illegible] - " " " " " " " 7000X

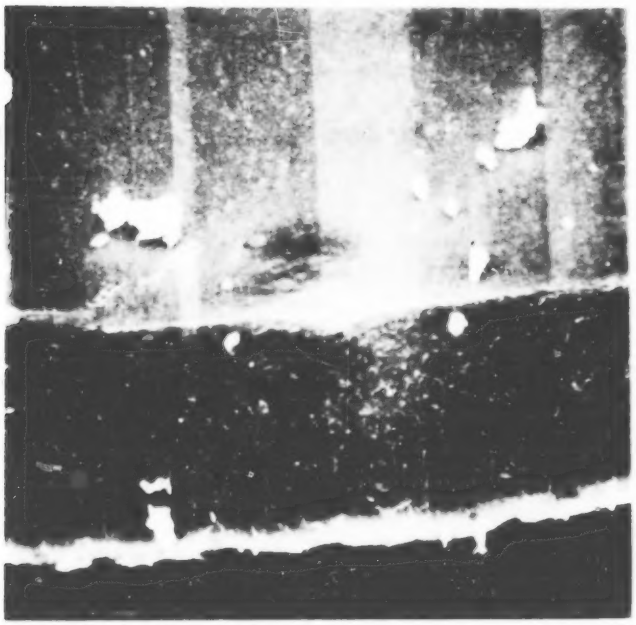


Fig. 66

Fig. 67

Fig. 68

Fig. 69

AUTOMOTIVE REFINISH

Figure 70 - 1000 Hours Exposure to 0.1 ppm O_3 - Unshaded - 700X

Figure 71 - " " " " 1.0 " " " - 700X

Figure 72 - " " " " 1.0 " SO_2 " - 700X

Figure 73 - " " " " 1.0 " " " - 7000X

Fig. 70

Fig. 71

Fig. 72

Fig. 73

SUMMARY AND CONCLUSIONS

In this study, research was concerned primarily with the development of test methods that are sufficiently sensitive to detect the initial degradation of selected exterior coatings exposed to various pollutant containing environments. The test methods employed in generating dose response data included erosion rates, attenuated total reflectance (ATR), gloss and sheen, surface roughness, tensile strength and scanning election microscopy. Five commerically important paint systems were selected for characterization including an oil house paint, latex coating, an alkyd industrial maintenance coating, a coil coating and an automotive refinish lacquer. The properties of the coating systems were determined periodically consequent to "short term" exposure at four exterior locations and to various pollutant containing environments under controlled, but accelerated conditions in a Xeonon light, Dew Cycle Weather-Ometer. The test sites for the former "short term" exterior study were located at Leeds (north central), North Dakota; Los Angeles, California; Chicago (Research Center), Illinois and Valparaiso, Indiana. These sites represent a "clean" rural environment, a high O_3 environment, a high SO_2 environment, and a relatively high O_3 environment plus a moderate SO_2 environment, respectively. In the accelerated laboratory exposure study, five environmental pollutant conditions were employed in the Weather-Ometer including a zero pollutant, 0.1 ppm and 1.0 ppm SO_2 and 0.1 ppm or 1.0 ppm O_3 level. The zero pollutant condition represents the control (clean air) with 0.1 ppm of each pollutant type considered representative of the levels frequently reached in polluted cities. The 1.0 ppm levels represent a highly polluted (industrial) site.

Concurrent to implementing the above two phases of the experimental program, a limited effort was devoted to reviewing existing exterior exposure records to establish the service life for each coating exposed in at least four locations, but not necessarily the same sites in all cases. The data from the latter study coupled with the results generated in the other two phases would potentially provide the required information for determining the reliability of any given test method to predict the service life of the selected coatings at each of the exposure sites. Unfortunately, the limited time element of this research contract did not permit the development of sufficient data for this extrapolation. It is also recognized, although not a specific requirement in satisfying the project objectives, that erosion rate curves established from the Weather-Ometer studies along with similar erosion data collected over an extended exterior exposure period (2 years minimum) can be used to provide an assessment of the economic loss attributable to the damage caused by atmospheric pollutants on coatings.

Conclusions from this investigation are:

(1) Erosion rate studies provide a definitive technique for determining the effect of atmospheric pollutants on the performance of exterior coatings. Consistent trends in erosion data were observed between the "short term" and Weather-Ometer exposure studies. ATR measurements on the coatings correlate well with the results of the erosion studies despite the generally mediocre spectra produced from the massive infrared absorption by the prime pigments. In addition, ATR analyses provide meaningful information concerning the chemical changes which occur in the film that would not normally be detected

with erosion studies. Scanning electron micrographs lend credence to erosion data and provide a permanent record of the topography changes which occur during exposure to aggressive environments.

The remaining three test methods employed in coating characterization provided only moderately useful information. Tensile strength measurements performed on the coatings exposed in the Weather-Ometer were virtually meaningless due to the rapid development of coating embrittlement. However, the tensile strength of coatings in the less severe exterior environment are following expected trends. Surface roughness analyses were even less definitive in differentiating between the effect of pollutant type and level on coating performance than tensile strength measurements. Gloss and sheen determinations are relevant only for the coatings which exhibit a high initial gloss level and/or a gradual decrease in gloss during exposure. The latter method of testing was capable of differentiating generally between the extremes in exterior exposure conditions.

(2) A significant conclusion derived particularly from the erosion, ATR and SEM analyses of the Weather-Ometer exposed panels is that atmospheric pollutants at levels encountered in polluted industrial sites (1.0 ppm SO_2 or O_3) and under the environmental conditions employed (temperature, relative humidity) exert a definite adverse effect on the performance of specific coatings compared to the zero pollutant condition. In general 1.0 ppm SO_2 as compared to the zero pollutant level caused a considerable effect on the oil house paint, a moderate effect on the latex and coil coating, but essentially no effect on the alkyd industrial maintenance coating and automotive refinish. Based on the comparison of erosion rates (slope, SO_2 at the 1 ppm affected the majority of coatings to a greater extent in either the shaded or unshaded condition than exposure to 1 ppm O_3. In addition, the ranking of a coating in terms of erosion rates (mils loss) is virtually independent of exposure to a pollutant type or level.

(3) Graphs of the slope of erosion rate with accompanying 95 percent confidence limits versus pollutant level (zero, 0.1 and 1 ppm SO_2 or O_3) for the Weather-Ometer exposed coatings support the a priori hypothesis that erosion rates are linearly related to pollutant concentration. Consequently, these curves along with exterior erosion data collected over an extended exposure period (two years minimum) provide the basis for assessing the economic loss or damage caused by atmospheric pollutants on coatings.

(4) The trends in erosion data for the "short term" exterior study after 7 months of exposure show that the mils loss of coating is higher at Chicago and Valparaiso than at the Los Angeles and North Dakota locations. These results were explained by the higher SO_2 levels at the former two exposure sites. ATR measurements and the SEM micrographs also supported this contention. It is also significant from the standpoint of correlation that the ranking in terms of mils loss for the exterior exposed coatings was virtually the same as the erosion rates generated in the accelerated studies regardless of exposure location (both north or south).

RECOMMENDATIONS FOR FURTHER RESEARCH

In view of the results generated in this experimental program and, considering that other forms of atmospheric pollutants, in addition to gaseous, may also exert a detectable adverse effect on coating performance the following areas for investigation are proposed:

1. Continuation of the "short term" exposure of selected coatings at the four exterior locations to confirm existing trends in detectable initial film damage data. Our considerable work with erosion studies shows that the exterior erosion rates of the selected coatings become linear after 9 months of exposure. Consequently, early erosion rates can be used effectively to predict long term durability as well as to provide a correlation factor for the linear erosion rates generated in Weather-Ometer exposures.

2. Repeat of the accelerated laboratory exposure study to simulate the detectable initial film damage encountered in exterior exposures using identical pollutant levels (i.e., zero, 0.1 and 1 ppm SO_2 or O_3), but at a lower relative humidity than in the present study. This effort would show whether the observed effect of the pollutants such as SO_2 or O_3 are moisture dependent.

3. Conduct a feasibility study concerned with determining the effect of atmospheric particulate matter on coating performance. This proposed area of study would be difficult to handle on an accelerated basis in the laboratory since there is no standard type of particulate matter nor well developed methods for controlling the level of particulates. However, this research effort could be accomplished by constructing a modified test fence at one of the four exterior test sites. Equipment is available for providing accelerated exposure but under exterior weather conditions typical for the given test site.

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